

van der Waals energy and pressure in dissipative media: Fluctuational electrodynamics and mode summation

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We derive the van der Waals energy and pressure in a planar multilayer system with arbitrary number of dissipative films between two half-spaces. A unique feature of this work is that the entire analysis is performed on the real frequency axis instead of summation over Matsubara frequencies on the imaginary frequency axis. The expression we obtain for van der Waals energy is a generalization of van Kampen and Schram's result for dissipationless media. By considering a specific case of a vacuum gap between multilayer objects with dissipative films, we show that the van der Waals energy due to the vacuum gap can not be interpreted simply as a sum of free energy of normal modes.

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The phenomena of adhesion and cohesion play an important role in many areas of science and technology; they are responsible for stiction-related failure in microelectromechanical devices [1]; microbial adhesion is responsible for the formation of biofilms [2], and they contribute to friction and wear between surfaces [3]. Adhesion and cohesion can be loosely defined as the molecular attraction that holds together surfaces of two different substances or two identical substances, respectively. Of the interactions that are responsible for adhesion and cohesion, the van der Waals or Casimir (we will use the term “van der Waals” to refer to both van der Waals and Casimir types of interactions from now on) interaction is universal and exists between all types of atoms as well as macroscopic objects.

Lifshitz [4] and Casimir [5] determined the force between two half-spaces as a function of separation using two seemingly different methods. Lifshitz relied on Rytov's theory of fluctuational electrodynamics [6] to determine the van der Waals pressure in the vacuum cavity between two half-spaces with frequency-dependent dielectric functions by evaluating the average electromagnetic stress tensor in vacuum due to thermal and zero-point fluctuations. Casimir evaluated the energy due to zero-point modes within a vacuum cavity between two parallel perfect electric conductors. The total free energy within this cavity is given by

$$E_c = k_B T \sum_n \ln \left[2 \sinh \left(\frac{\hbar \omega_n}{2k_B T} \right) \right], \quad (1)$$

where each value of n corresponds to a different mode, k_B is the Boltzmann constant, $2\pi\hbar$ is the Planck constant, and T is the absolute temperature of the system. The force arises from the variation of the total free energy with thickness of the vacuum cavity. van Kampen *et al.* [7] and Schram [8] were the first to derive the Lifshitz formula for van der Waals pressure starting from summation of energy of electromagnetic modes in the vacuum cavity when the two half-spaces are dispersive but not dissipative. A good summary of the similarities and

differences between the fluctuational electrodynamics method and the mode-summation method can be found in Ref. [9].

What happens to the van der Waals pressure when the vacuum gap is filled with a dissipative material? While Lifshitz' original method can not be utilized directly because the stress tensor for arbitrary electromagnetic fields is not defined in dissipative media, Dzyaloshinskii, Lifshitz, and Pitaevskii [10] used techniques from quantum field theory to determine the van der Waals pressure in dissipative media. Barash and Ginzburg [11,12] justified the usage of Eq. (1) even in dissipative media on the grounds that it is possible to ascribe thermodynamic functions to electromagnetic fields in equilibrium with matter [12,13]. We derived a first-principles method, without using quantum field theory, of determining the van der Waals pressure in a dissipative and dispersive film within a multilayer structure by calculating the Maxwell stress tensor in fictitious layers of vacuum introduced in the structure [14]. Since the fictitious vacuum layers are eventually made to vanish, we retrieve the original system of interest. Using this method, the expression for van der Waals pressure in a dissipative film was shown to agree exactly with that obtained by Dzyaloshinskii, Lifshitz, and Pitaevskii [10]. Despite many works on the mode-summation method, determining van der Waals energy when at least one of the materials is dissipative is still a topic of active research [9,15–20] and has not been resolved entirely.

We, perhaps flippantly, remarked in Ref. [14] that the extension of our theoretical formalism to multilayer systems is simply an exercise in determining the appropriate Fresnel reflection and transmission coefficients. While there is some truth to that statement, as we will show here, it also underplays what can be learned from a complete analysis of the multilayer problem. The primary contribution of this paper is a derivation of an expression for van der Waals free energy of a planar multilayer system consisting of N films with dissipative and dispersive dielectric functions and magnetic permeabilities between two half-spaces. The problem of van der Waals pressure and energy in planar multilayer systems has been considered before by many researchers. A subset of those works, that we are familiar with, is referenced here [21–26]. A unique feature of our derivation is that, even though it involves finding the pressure and energy in a dissipative

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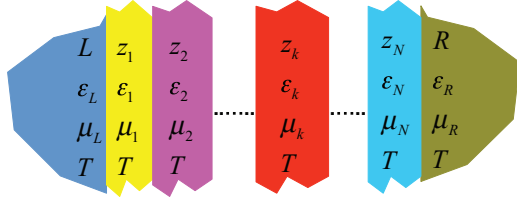


FIG. 1. (Color online) Schematic of a multilayer system between two half-spaces L and R .

material, it proceeds entirely along the real frequency axis.¹ Performing the analysis along the real frequency axis has (at least) three advantages over the analysis along the imaginary axis: (1) spectral contributions to the energy and pressure can be obtained [27], (2) contributions can be divided into propagating and evanescent waves [27–29], and (3) determination of thermal nonequilibrium van der Waals energy and pressure in a dissipative material, if at all possible to determine, will require analysis along the real frequency axis since the functions to be integrated are no longer analytic in the upper half of the complex frequency plane [30,31]. The other important contribution is to show that the van der Waals energy can not be expressed exclusively as a sum of free energy of normal modes when any of the materials is dissipative.

This paper is structured as follows: Expressions for van der Waals pressure and energy in a thin film as obtained from Lifshitz theory [4] and the generalization by Dzyaloshinskii, Lifshitz, and Pitaevskii [10,14] are given in Sec. I. In Sec. II, expressions for van der Waals energy of N -layered system with dissipative materials are obtained using fluctuational electrodynamics and the principle of conservation of energy. In Sec. III, we illuminate the similarities and differences between the fluctuational electrodynamics method and the mode-summation method to evaluate van der Waals energy when dissipation is present. We discuss the implications of our work in Sec. IV and outline some issues we have not been able to resolve to our satisfaction. We summarize our contributions in Sec. V.

I. LIFSHITZ THEORY OF VAN DER WAALS ENERGY AND PRESSURE IN A PLANAR FILM

Consider a planar multilayer stack of N films sandwiched between two half-spaces L and R (see Fig. 1). All objects are at the same temperature T . The films are characterized by thickness z_k , dielectric permittivity ϵ_k , and magnetic permeability μ_k , where $k \in \{1, 2, \dots, N\}$. When k is replaced by L or R , it refers to the properties of half-spaces L or R . All dielectric permittivities and magnetic permeabilities are frequency dependent. At finite temperature, evaluation of van der Waals energy involves integration of functions over the real frequency axis or summation of an infinite sequence, each term of which is evaluated at a Matsubara frequency $i\xi_n = in2\pi k_B T / \hbar$, $n = 0, 1, 2, \dots$. The integrals along the frequency

axis are of the form $\int_{0^+}^{\infty} d\omega \coth(\frac{\hbar\omega}{2k_B T}) \text{Im} f(\omega)$, where $\text{Im} f$ stands for the imaginary part of f , and $f(\omega + i\xi)$ is an analytic function with no poles in the upper half complex frequency plane ($\xi > 0$), and the integral $\int_{0^+}^{\infty} d\omega \equiv \lim_{\delta \rightarrow 0} \int_{|\delta|}^{\infty} d\omega$. $f(\omega)$ also satisfies the property that $f(-\omega) = f^*(\omega)$. The analyticity of f can be exploited to replace the integral by a more (computationally) convenient sum $-\frac{2\pi k_B T}{\hbar} \sum_{n=0}^{\infty} f(i\xi_n)$. $\sum_{n=0}^{\infty}$ indicates that the $n = 0$ term is multiplied by $\frac{1}{2}$ [4]. Both approaches yield identical values of van der Waals energy and pressure. In this paper, though, all mathematical entities are defined along the real frequency axis. Computations of integrals are also done along the real frequency axis.

The integral expression for van der Waals pressure in the k th layer, i.e., that part of the fluctuational pressure that is influenced by the presence of discontinuities in electrical permittivity or magnetic permeability, is given by² [14]

$$p_k(z_k) = - \sum_{p=e,h} \int_0^{\infty} \frac{dk_{\rho} k_{\rho}}{2\pi} \int_{0^+}^{\infty} d\omega \frac{\hbar}{\pi} \coth \left[\frac{\hbar\omega}{2k_B T} \right] \times \text{Im} \left[ik_{z_k} \frac{\tilde{R}_{k,[k-1]}^{(p)}(\omega) \tilde{R}_{k,[k+1]}^{(p)}(\omega) e^{i2k_{z_k} z_k}}{1 - \tilde{R}_{k,[k-1]}^{(p)}(\omega) \tilde{R}_{k,[k+1]}^{(p)}(\omega) e^{i2k_{z_k} z_k}} \right], \quad (2)$$

where z_k is the thickness of the k th layer, k_{ρ} is the magnitude of the in-plane wave vector, $k_{z_k}(\omega) = \sqrt{\frac{\omega^2}{c^2} \epsilon_k(\omega) \mu_k(\omega) - k_{\rho}^2}$ is the z component of wave vector in the k th layer, and $\tilde{R}_{k,[k-1]}^{(p)}$ is the generalized Fresnel reflection coefficients for p -polarized plane waves from film k incident at the interface with the multilayer to the left of k (composed of half-space L and films 1 through $k-1$). Similarly, $\tilde{R}_{k,[k+1]}^{(p)}$ is the generalized Fresnel reflection coefficient for p -polarized plane waves from film k incident at the interface with the multilayer to the right (composed of films $k+1$ through N and half-space R). $p = e, h$ stand for transverse electric and transverse magnetic polarizations, respectively. $\tilde{R}_{k,[k\pm 1]}^{(p)}$ can be determined from the following recursion relation [32]:

$$\tilde{R}_{k,[k\pm 1]}^{(p)} = \frac{R_{k,k\pm 1}^{(p)} + \tilde{R}_{k\pm 1,[k\pm 2]}^{(p)} e^{i2k_{z(k\pm 1)} z_{k\pm 1}}}{1 + R_{k,k\pm 1}^{(p)} \tilde{R}_{k\pm 1,[k\pm 2]}^{(p)} e^{i2k_{z(k\pm 1)} z_{k\pm 1}}}, \quad (3)$$

where

$$R_{k,k\pm 1}^{(p)} = \frac{k_{z_k} / \alpha_k^{(p)} - k_{z(k\pm 1)} / \alpha_{k\pm 1}^{(p)}}{k_{z_k} / \alpha_k^{(p)} + k_{z(k\pm 1)} / \alpha_{k\pm 1}^{(p)}} \quad (4)$$

are Fresnel coefficients for p -polarized plane waves at the interface between layers of materials with properties ϵ_k, μ_k and $\epsilon_{k\pm 1}, \mu_{k\pm 1}$. $\alpha_k^{(p)} = \epsilon_k$ and μ_k for h and e polarized waves, respectively. Because of the proliferation of reflection coefficients in this paper, the different symbols are consolidated in Table I.

¹The transformation of the integral to summation of functions evaluated at Matsubara frequencies along the positive imaginary frequency axis, while not necessary, can be used eventually for computational expediency.

²Most works give the equivalent expression for van der Waals pressure obtained by series summation over the Matsubara frequencies.

TABLE I. List of reflection coefficients used in this paper. See the equation numbers (if provided) for the definition of the reflection coefficient.

Symbol	Equation	Comment
$R_{i,j}^{(p)}$	Eq. (4)	Reflection coefficient of a plane wave from half-space of material i incident at an interface with half-space of material j . $i, j = V \Rightarrow$ one of the half-spaces is vacuum.
$\tilde{R}_{k,[k-1]}^{(p)}$	Eq. (3)	Reflection coefficient of a plane wave in material k incident on the multilayer structure to the left of film k .
$\tilde{R}_{k,[k+1]}^{(p)}$	Eq. (3)	Reflection coefficient of a plane wave in material k incident on the multilayer structure to the right of film k .
$\tilde{R}_{V,k}^{(p)}$	Eq. (10)	Reflection coefficient of a plane wave from vacuum incident at an interface of a thin film of material k surrounded by vacuum on both sides.
$\tilde{R}_{V,[k-1]}^{(p)}$		Reflection coefficient of a plane wave in vacuum incident on the multilayer structure to the left of film $k - 1$ (including film $k - 1$).
$\tilde{R}_{V,[k+]}^{(p)}$		Reflection coefficient of a plane wave in vacuum incident on the multilayer structure to the right of film k (including film k).

$p_k(z_k)$ can be obtained by differentiation of $U_k(z_k)$ with respect to z_k , where $U_k(z_k)$ is given by

$$U_k(z_k) = \sum_{p=e,h} \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \int_{0^+}^\infty d\omega \frac{\hbar}{2\pi} \coth \left[\frac{\hbar\omega}{2k_B T} \right] \times \text{Im} \ln \left(1 - \tilde{R}_{k,[k-1]}^{(p)} \tilde{R}_{k,[k+1]}^{(p)} e^{i2k_\rho z_k} \right). \quad (5)$$

After integration by parts, Eq. (5) can be written as

$$U_k(z_k) = - \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \int_{0^+}^\infty d\omega \frac{k_B T}{\pi} \ln \left[2 \sinh \frac{\hbar\omega}{2k_B T} \right] \times \sum_{p=e,h} \text{Im} \frac{\partial}{\partial \omega} \ln \left(1 - \tilde{R}_{k,[k-1]}^{(p)} \tilde{R}_{k,[k+1]}^{(p)} e^{i2k_\rho z_k} \right). \quad (6)$$

Although they do not appear as arguments of the function, p_k and U_k are in fact implicit functions of $z_1, z_2, \dots, z_{k-1}, z_{k+1}, \dots, z_N$ as well as ε, μ of all the materials. U_k , however, can not be interpreted as the van der Waals energy of the entire multilayer system since $U_m(z_m) \neq U_n(z_n)$ if $m \neq n$ [25].

The question we ask here is as follows: what is the energy $U_{LR}^{(N)}$, which is a function of properties and dimensions of all materials in the multilayer system, from which the pressure in any constituent film can be obtained through the relation

$$p_k(z_k) = - \frac{\partial U_{LR}^{(N)}}{\partial z_k}, \quad k = 1, 2, \dots, N. \quad (7)$$

Equation (7) can be interpreted as a system of N first-order partial differential equations, the solution of which yields $U_{LR}^{(N)}$. Instead of trying to solve Eq. (7) directly, we will use conservation of energy to construct $U_{LR}^{(N)}$.

II. VAN DER WAALS ENERGY OF A PLANAR MULTILAYER SYSTEM

The method outlined in Ref. [14] can be summarized as a simple principle: the free energy of a planar multilayer system can be obtained by adding to the free energy of each component (half-spaces L and R , and N films) the work done in assembling them into the desired multilayer system. The

mathematical statement of this principle is given by

$$U_{LR}^{(N)} = U_{LV}^{(0)} + \sum_{k=1}^N [U_{VV}^{(1)}(z_k) + W_k(z_1, \dots, z_k)] + U_{VR}^{(0)} + W_R(z_1, \dots, z_N), \quad (8)$$

where we use the following notation: the free energy of N films bounded by half-spaces L and R is represented by $U_{LR}^{(N)}$; $U_{VV}^{(1)}(z_k)$ stands for the free energy of a thin film of thickness z_k surrounded by vacuum either side; $W_k(z_1, \dots, z_{k-1}, z_k)$ stands for the work done against the van der Waals pressure in vacuum in order to translate a free-standing thin film of thickness z_k from infinite separation to the surface of the multilayer system formed by $k - 1$ contiguous films bounded by half-space L and vacuum on either side; $W_R(z_1, \dots, z_N)$ is the work done in translating half-space R from infinite separation to the surface of the rest of the multilayer structure in Fig. 1; $U_{LV}^{(0)}$ ($U_{VR}^{(0)}$) is the free energy of a half-space of L (R) adjacent to vacuum and is a constant. $U_{LV}^{(0)}$ and $U_{VR}^{(0)}$ do not play any role in determining the van der Waals pressure in any of the thin films. It is worth noting that the partial sum $U_{LV}^{(0)} + \sum_{k=1}^j [U_{VV}^{(1)}(z_k) + W_k(z_1, \dots, z_k)]$ is in fact $U_{LV}^{(j)}$, the free energy of the multilayer system formed by the first j films sandwiched between half-space L and vacuum.

Apart from $U_{LV}^{(0)}$ and $U_{VR}^{(0)}$,³ the terms in the right-hand side of Eq. (8) can be written in terms of appropriate generalized Fresnel reflection coefficients. $U_{VV}^{(1)}(z_k)$ can be written as [14]

$$U_{VV}^{(1)}(z_k) = \sum_{p=e,h} \lim_{\Delta_k \rightarrow 0} \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \int_{0^+}^\infty d\omega \coth \left[\frac{\hbar\omega}{2k_B T} \right] \times \frac{\hbar}{2\pi} \text{Im} \ln \left(1 - R_{V,k}^{(p)} \tilde{R}_{V,k}^{(p)} e^{i2k_\rho \Delta_k} \right), \quad (9)$$

³ $U_{LV}^{(0)}$ and $U_{VR}^{(0)}$ can also be written in terms of Fresnel reflection coefficients but only with the inclusion of a parameter D_0 [37] that is proportional to the minimum separation between two atomic planes of the constituent material.

where $k_{zv}(\omega) = \sqrt{\frac{\omega^2}{c^2} - k_\rho^2}$, and

$$\tilde{R}_{V,k}^{(p)} = \frac{R_{V,k}^{(p)}(1 - e^{i2k_{zk}z_k})}{1 - R_{V,k}^{(p)2}e^{i2k_{zk}z_k}} \quad (10)$$

is the reflection coefficient of a plane wave in vacuum incident at the interface with a film of material k surrounded by vacuum on both sides. Although the integral in Eq. (9) has a singularity at $\Delta_k = 0$, it does not pose a problem when computing $\partial U_{VV}(z_k)/\partial z_k$ [see the discussion following Eq. (14) for an explanation; also see Ref. [14]]. $W_k(z_1, \dots, z_k)$ is given by

$$W_k(z_1, \dots, z_k) = - \sum_{p=e,h} \lim_{\delta_k \rightarrow 0} \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \int_{0^+}^\infty d\omega \frac{\hbar}{2\pi} \coth \left[\frac{\hbar\omega}{2k_B T} \right] \text{Im} \ln \left(1 - \tilde{R}_{V,[k-1]}^{(p)} \tilde{R}_{V,k}^{(p)} e^{i2k_{zv}\delta_k} \right), \quad (11)$$

where $\tilde{R}_{V,[k-1]}^{(p)}$ is the generalized Fresnel reflection coefficient for a wave in vacuum incident at the interface between vacuum and the $(k-1)$ th film. $k-2$ other films are present between the $(k-1)$ th film and the half-space L . $\tilde{R}_{V,[k-1]}^{(p)}$ can be determined by using the recursion relation in Eq. (3). An important feature of Eqs. (9) and (11) is that both of them are obtained by calculating the Maxwell stress tensor only in vacuum, where the stress tensor is defined unambiguously [14].

Substituting Eqs. (9) and (11) into Eq. (8), $U_{LR}^{(N)}$ is given by

$$U_{LR}^{(N)} = \sum_{p=e,h} \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \int_{0^+}^\infty d\omega \frac{\hbar}{2\pi} \coth \left[\frac{\hbar\omega}{2k_B T} \right] \left\{ \lim_{\delta_1 \rightarrow 0} \text{Im} \ln \left[1 + R_{1,V}^{(p)} \tilde{R}_{V,L}^{(p)} e^{i2k_{zv}\delta_1} \right] + \sum_{k=1}^N \lim_{\substack{\delta_{k+1} \rightarrow 0 \\ \Delta_k \rightarrow 0}} \text{Im} \ln \left[\frac{(1 + R_{V,k}^{(p)} \tilde{R}_{k,[k-1]}^{(p)} e^{i2k_{zk}z_k})(1 + R_{k+1,V}^{(p)} \tilde{R}_{V,[k]}^{(p)} e^{i2k_{zv}\delta_{k+1}})}{1 - R_{V,k}^{(p)2} e^{i2k_{zv}\Delta_k}} \right] \right\}. \quad (12)$$

It can be further simplified by realizing that

$$\begin{aligned} & (1 + R_{V,k}^{(p)} \tilde{R}_{k,[k-1]}^{(p)} e^{i2k_{zk}z_k})(1 + R_{k+1,V}^{(p)} \tilde{R}_{V,[k]}^{(p)} e^{i2k_{zv}\delta_{k+1}}) \\ &= (1 + R_{V,k}^{(p)} \tilde{R}_{k,[k-1]}^{(p)} e^{i2k_{zk}z_k}) \left(1 + R_{k+1,V}^{(p)} \frac{R_{V,k}^{(p)} + \tilde{R}_{k,[k-1]}^{(p)} e^{i2k_{zk}z_k}}{1 + R_{V,k}^{(p)} \tilde{R}_{k,[k-1]}^{(p)} e^{i2k_{zk}z_k}} e^{i2k_{zv}\delta_{k+1}} \right) \\ &= (1 + R_{k+1,V}^{(p)} R_{V,k}^{(p)} e^{i2k_{zv}\delta_{k+1}}) \left(1 - \frac{R_{k,V}^{(p)} + R_{V,k+1}^{(p)} e^{i2k_{zv}\delta_{k+1}}}{1 + R_{k,V}^{(p)} R_{V,k+1}^{(p)} e^{i2k_{zv}\delta_{k+1}}} \tilde{R}_{k,[k-1]}^{(p)} e^{i2k_{zk}z_k} \right) \\ &= (1 + R_{k+1,V}^{(p)} R_{V,k}^{(p)} e^{i2k_{zv}\delta_{k+1}}) (1 - R_{k,k+1}^{(p)} \tilde{R}_{k,[k-1]}^{(p)} e^{i2k_{zk}z_k}). \end{aligned} \quad (13)$$

In going from the last-but-one equation to the last line in Eq. (13), we have used the fact that we will eventually take the limit as $\delta_{k+1} \rightarrow 0$ [see Eqs. (12) or (14)]. Using Eq. (13), Eq. (12) can be simplified to

$$\begin{aligned} U_{LR}^{(N)} &= - \sum_{p=e,h} \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \int_{0^+}^\infty d\omega \frac{\hbar}{2\pi} \coth \left[\frac{\hbar\omega}{2k_B T} \right] \\ &\quad \times \text{Im} \left\{ \lim_{\delta_k \rightarrow 0} \sum_{k=0}^N \ln (1 - R_{V,k+1}^{(p)} R_{V,k}^{(p)} e^{i2k_{zv}\delta_{k+1}}) - \lim_{\Delta_k \rightarrow 0} \sum_{k=1}^N \ln (1 - R_{V,k}^{(p)2} e^{i2k_{zv}\Delta_k}) + \sum_{k=1}^N \ln (1 - R_{k,k+1}^{(p)} \tilde{R}_{k,[k-1]}^{(p)} e^{i2k_{zk}z_k}) \right\} \\ &= C - \sum_{p=e,h} \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \int_{0^+}^\infty d\omega \frac{\hbar}{4\pi^2} \coth \left[\frac{\hbar\omega}{2k_B T} \right] \text{Im} \ln \prod_{k=1}^N (1 - R_{k,k+1}^{(p)} \tilde{R}_{k,[k-1]}^{(p)} e^{i2k_{zk}z_k}), \end{aligned} \quad (14)$$

where $C \equiv C(\delta_1, \dots, \delta_{N+1}; \Delta_1, \dots, \Delta_N)$ is independent of all z_k ($k = 1, \dots, N$). In Eq. (14), $R_{N,N+1} \equiv R_{N,R}$, $R_{V,0} \equiv R_{V,L}$, $R_{V,N+1} \equiv R_{V,R}$, and $\tilde{R}_{1,[0]} \equiv \tilde{R}_{1,L}$. Only the second term contributes to van der Waals pressure in any of the layers. The implication of Eq. (14) is that the van der Waals energy of any planar multilayer system between two half-spaces can be split into two parts: a configuration-dependent part that contributes to van der Waals pressure, and a singular part that does not.

The modes themselves are obtained by setting the argument of the logarithm function in Eq. (14) to zero, i.e., the

dispersion relation for the N -layer structure in Fig. 1 is given by $\tilde{D}^{(p)}(\omega, k_\rho) = \prod_{k=1}^N (1 - R_{k,k+1}^{(p)} \tilde{R}_{k,[k-1]}^{(p)} e^{i2k_{zk}z_k}) = 0$. Hence, the finite z_k -dependent part of the van der Waals energy in Eq. (14) can also be written as

$$U_{LR}^{(N)} = - \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \int_{0^+}^\infty d\omega \frac{k_B T}{\pi} \ln \left[2 \sinh \frac{\hbar\omega}{2k_B T} \right] \times \sum_{p=e,h} \frac{\partial}{\partial \omega} \text{Im} \ln \tilde{D}^{(p)}(\omega, k_\rho). \quad (15)$$

Equation (15) for $U_{LR}^{(N)}$ is a generalization of van Kampen's and Schram's expression for van der Waals energy of an N -layer medium at finite temperature [7,8,18,19,33]. However, two important features need to be kept in mind: (1) Eqs. (14) and (15) are obtained without having to rely on the assumption that computation of van der Waals free energy needs the summation of free energies of all fundamental modes of the system, and (2) unlike in all the literature regarding the mode-summation method we are aware of [7–9,16,18–20,33], the layer in which the van der Waals pressure is to be calculated can have dissipative properties.

III. EXCURSIONS INTO THE LOWER HALF PLANE IN THE PRESENCE OF DISSIPATION

Since Eq. (15) is a generalization of van Kampen's and Schram's formula for van der Waals energy to the case of dissipative materials, it is natural to ask if we can also express it as a sum over normal modes, thereby lending legitimacy to the idea of mode summation in dissipative media. Specifically, we are interested in the van der Waals pressure in a vacuum gap between two multilayer objects which contain dissipative thin films. To show the relation between Eq. (15) and the sum of mode energies, the integration path along the real axis should be completed into a closed contour so as to include all the normal modes that contribute to van der Waals energy, which lie in the lower half of the complex frequency plane if at least one of the materials in the multilayer system is dissipative.

The vacuum gap of length l_v in which the van der Waals pressure is to be determined is in-between multilayer objects marked 1 and 2 in Fig. 2. To simplify the analysis, the multilayer structure is placed inside a cavity with a perfect reflector at either end, marked "Schram's perfect reflector" (SPR) in Fig. 2. This is done in order to eliminate branch points corresponding to the half-spaces L and R that would have otherwise been present [8]. In addition to the usual SPR employed by many, we added a layer of dissipative material to the surface of SPR to create "Schram's imperfect reflector" (SIR in Fig. 2). In particular, SPR to the left has a coating of

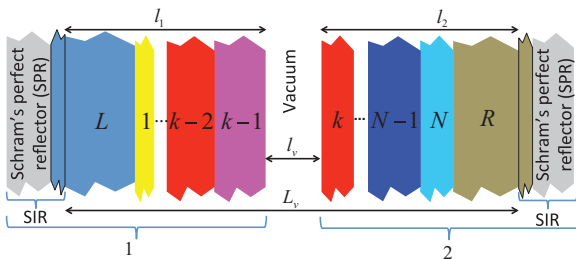


FIG. 2. (Color online) The multilayer structure is sandwiched between two perfect reflectors (marked as "Schram's perfect reflector") in order to eliminate branch points corresponding to the half-spaces L and R that would have otherwise been present at either end. A vacuum gap of length l_v is introduced between the films $k-1$ and k . The van der Waals energy of the multilayer system due to the presence of the vacuum gap is to be interpreted as a sum over normal mode free energies.

material L and that to the right has a coating of material R . The dissipative layers, the thicknesses of which are immaterial, are present to ensure that the frequencies of normal modes of the two SIRs, with only vacuum in-between, are pushed from the real axis to the lower half of the complex frequency plane. If the materials L and R are vacuum, infinitesimal dissipation is added. The total thickness of multilayer object 1 is l_1 and that of multilayer object 2 is l_2 . $L_v = l_v + l_1 + l_2$ is the gap between the two SIRs. Eventually the thickness z_L and z_R of L and R , respectively, and hence l_1 and l_2 , are made to approach ∞ to recreate the multilayer system of interest. The assumption of vacuum cavity is not unduly restrictive as we have shown in Sec. II that each term in the right-hand side of Eq. (8) corresponds to the van der Waals energy of a multilayer system similar to the one shown in Fig. 2 (albeit without the reflectors at either end). Hence, if we can express the van der Waals energy of the vacuum cavity in Fig. 2 in terms of normal modes, then we can use the theory in Sec. II to express the energy of any planar multilayer system in terms of energy of normal modes.

The regularized van der Waals energy of the multilayers at a vacuum gap l is the difference between (1) the work required to translate the two multilayer objects, including the reflectors, from $l_v \rightarrow \infty$ to $l_v = l$, and (2) the work done in translating the two SIRs, with only vacuum in-between, from $L_v \rightarrow \infty$ to $L_v = l + l_1 + l_2$. Denoting this van der Waals energy as $U_v(l)$, and by using Eq. (14), we can write

$$U_v(l) = \sum_{p=e,h} \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \int_{0^+}^\infty d\omega \frac{\hbar}{2\pi} \coth \left[\frac{\hbar\omega}{2k_B T} \right] \times \left[\text{Im} \left[\lim_{l_v \rightarrow \infty} \ln \left(\frac{D_v^{(p)}(\omega, k_\rho; l)}{D_v^{(p)}(\omega, k_\rho; l_v)} \right) \right] - \text{Im} \left[\lim_{L_v \rightarrow \infty} \ln \left(\frac{D_{\text{SIR}}^{(p)}(\omega, k_\rho; l + l_1 + l_2)}{D_{\text{SIR}}^{(p)}(\omega, k_\rho; L_v)} \right) \right] \right]. \quad (16)$$

In Eq. (16), $D_v^{(p)}(\omega, k_\rho; l) = 1 - \tilde{R}_{V,[k-1]}^{(p)} \tilde{R}_{V,[k+]}^{(p)} e^{i2k_z l}$ is the dispersion relation for modes in the entire structure confined between the two SPRs. $\tilde{R}_{V,[k-1]}^{(p)}$ and $\tilde{R}_{V,[k+]}^{(p)}$ are the reflection coefficients of multilayer systems 1 and 2 inclusive of the corresponding SIR. As $\omega z_L/c \rightarrow \infty$, $\tilde{R}_{V,[k-1]}^{(p)} \rightarrow \tilde{R}_{V,[k-1]}^{(p)}$. Similarly, as $\omega z_R/c \rightarrow \infty$, $\tilde{R}_{V,[k+]}^{(p)} \rightarrow \tilde{R}_{V,[k+]}^{(p)}$. $D_{\text{SIR}}^{(p)}(\omega, k_\rho; l + l_1 + l_2) = 1 - \tilde{R}_{V,L}^{(p),\text{SIR}} \tilde{R}_{V,R}^{(p),\text{SIR}} e^{i2k_z(l+l_1+l_2)}$ is the dispersion relation for the cavity between the two SIRs with the space between them filled by vacuum, where $\tilde{R}_{V,L}^{(p),\text{SIR}}$ and $\tilde{R}_{V,R}^{(p),\text{SIR}}$ are reflection coefficients of SIR to the left and right, respectively. The integral from $\omega = 0^+$ to $\omega = \infty$ can be rewritten as an integral over the entire real axis by appropriately defining the integrand along the negative real axis. We know that $\varepsilon(-|\omega|) = \varepsilon^*(|\omega|)$. The z direction wave vectors k_{zj} obey the relation $k_{zj}(-|\omega|) = -k_{zj}^*(|\omega|)$ for all j (the behavior of the wave vector in vacuum is discussed specifically in Sec. III A). With this definition, we can see that $\ln D_v^{(p)}(\omega, k_\rho; l)$ evaluated at $|\omega|$ and $-|\omega|$ are complex conjugates of each other. To see how Eq. (16) can be written as a sum over mode energies, let us rewrite Eq. (16) as an integral along the entire real frequency

axis:

$$U_v(l) = \sum_{\rho=e,h} \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \int_{-\infty}^\infty d\omega \frac{i\hbar}{4\pi} \coth \left[\frac{\hbar\omega}{2k_B T} \right] \times \left[\left[\lim_{l_v \rightarrow \infty} \ln \left(\frac{D_v^{(p)}(\omega, k_\rho; l)}{D_v^{(p)}(\omega, k_\rho; l_v)} \right) \right] - \left[\lim_{L_v \rightarrow \infty} \ln \left(\frac{D_{\text{SIR}}^{(p)}(\omega, k_\rho; l + l_1 + l_2)}{D_{\text{SIR}}^{(p)}(\omega, k_\rho; L_v)} \right) \right] \right]. \quad (17)$$

$\int_{-\infty}^\infty d\omega$ should be interpreted as $\lim_{\delta \rightarrow 0} \int_{-\infty}^{-|\delta|} d\omega + \lim_{\delta \rightarrow 0} \int_{|\delta|}^\infty d\omega$. When considered as an integral over ω alone, i.e., the integral over k_ρ is performed first, there should be no difference in evaluating Eq. (15) in the upper side ($\omega + i0$) or lower side ($\omega - i0$) of the real frequency axis. However, when the order of integration is changed, i.e., integration over ω is done first, branch points arising due to the square root in the definition of the z -component wave vector $[k_{zj}(\omega)]$ complicate matters. The next section addresses questions related to the behavior of $\lim_{l_v \rightarrow \infty} \ln \left[\frac{D_v^{(p)}(\omega, k_\rho; l)}{D_v^{(p)}(\omega, k_\rho; l_v)} \right]$ at different segments of the real frequency axis in the upper and lower half planes.

A. Branch cuts and contours of integration in upper and lower halves of complex frequency plane

Since $k_{zj}, j = L, R, 1, 2, \dots, k - 1, v, k, \dots, N$ are all relevant to the problem, it appears as though we have as many branch-point pairs as there are films. However, we will show that only the branch points corresponding to the vacuum region are important for evaluating $D_v^{(p)}(\omega, k_\rho; l)$. The complex frequencies corresponding to $k_{zj} = 0$ are valid branch points if we can show that the function $D_v^{(p)}(\omega, k_\rho; l)$ takes on different values when the sign of k_{zj} is changed. The effect of changing the sign of only k_{zj} , at the same ω is $R_{j, j\pm 1}^{(p)}(-k_{zj}) = R_{j, j\pm 1}^{(p)}(k_{zj})$. Keeping in mind that k_{zv} affects only $R_{V, k-1}$, the effect of changing the sign of only k_{zv} is $R_{V, k-1}^{(p)}(-k_{zv}) = R_{V, k-1}^{(p)}(k_{zv})$. Hence,

$$\begin{aligned} \tilde{R}_{V, [k-1]}^{(p)}(-k_{zv}) &= \frac{R_{V, k-1}^{(p)-1}(k_{zv}) + \tilde{R}_{k-1, [k-2]}^{(p)} e^{i2k_z(k-1)z_{k-1}}}{1 + R_{V, k-1}^{(p)-1}(k_{zv}) \tilde{R}_{k-1, [k-2]}^{(p)} e^{i2k_z(k-1)z_{k-1}}} \\ &= \tilde{R}_{V, [k-1]}^{(p)-1}(k_{zv}). \end{aligned} \quad (18)$$

Similarly, we can also show that $\tilde{R}_{V, [k+1]}^{(p)}(-k_{zv}) = \tilde{R}_{V, [k+1]}^{(p)-1}(k_{zv})$. Clearly, the frequencies at which $k_{zv} = 0$ are branch points for evaluating $D_v^{(p)}(\omega, k_\rho; l)$. Since $\tilde{R}_{k-1, [k-2]}^{(p)}(-k_{z(k-1)}) = \tilde{R}_{k-1, [k-2]}^{(p)-1}(k_{z(k-1)})$, we have $\tilde{R}_{V, [k-1]}^{(p)}(-k_{z(k-1)}) = \tilde{R}_{V, [k-1]}^{(p)}(k_{z(k-1)})$. Only $\tilde{R}_{k-1, [k-2]}^{(p)}$ depends on $k_{z(k-2)}$ or other wave vectors $k_{zj}, j = L, 1, \dots, k - 3$. By extension of Eq. (18), we see that $\tilde{R}_{k-1, [k-2]}^{(p)}(-k_{z(k-2)}) = \tilde{R}_{k-1, [k-2]}^{(p)}(k_{z(k-2)})$ and, therefore, $\tilde{R}_{V, [k-1]}^{(p)}(-k_{z(k-2)}) = \tilde{R}_{V, [k-1]}^{(p)}(k_{z(k-2)})$. This analysis can be extended to show that $\tilde{R}_{V, [k-1]}^{(p)}$ does not change when the sign of any of the wave vectors $k_{zj}, j = L, 1, \dots, k - 1$ is changed. The same argument holds true for $\tilde{R}_{V, [k+1]}^{(p)}$. Hence, the only branch points correspond to the frequencies at which $k_{zv} = 0$.

For a given value of k_ρ , these frequencies correspond to $\omega = \pm ck_\rho$. We draw branch cuts extending from ck_ρ to ∞ and from $-ck_\rho$ to $-\infty$. The presence of branch cuts implies that $D_v^{(p)}(\omega, k_\rho; l)$ takes on different values in the upper and lower half planes because of the changing sign of k_{zv} . k_{zv} is defined such that $\text{Im}k_{zv} \geq 0$ all over the complex frequency plane. In the upper half plane,

$$k_{zv}(\omega) = \begin{cases} |k_{zv}|, & \omega > ck_\rho \\ i|k_{zv}|, & |\omega| < ck_\rho \\ -|k_{zv}|, & \omega < -ck_\rho. \end{cases} \quad (19)$$

In the lower half plane,

$$k_{zv}(\omega) = \begin{cases} -|k_{zv}|, & \omega > ck_\rho \\ i|k_{zv}|, & |\omega| < ck_\rho \\ +|k_{zv}|, & \omega < -ck_\rho. \end{cases} \quad (20)$$

The definition of k_{zv} in Eqs. (19) and (20) is in agreement with the relationship between z -component wave vectors at $|\omega|$ and $-|\omega|$, which is given by $k_{zj}(-|\omega|) = -k_{zj}^*(|\omega|)$.

Using the definitions of k_{zv} , the values of $I^{(p)}(\omega, k_\rho) = \lim_{l_v \rightarrow \infty} \ln \left[\frac{D_v^{(p)}(\omega, k_\rho; l)}{D_v^{(p)}(\omega, k_\rho; l_v)} \right]$ in different segments of the real frequency axis in the lower half plane can be related to the corresponding values at appropriate positions in the upper half plane and positive frequencies. For $0 < \omega < ck_\rho$, the reflection coefficients $\tilde{R}_{V, [k-1]}^{(p)}$ and $\tilde{R}_{V, [k+1]}^{(p)}$, as well as $e^{i2k_{zv}l}$, are evaluated at $k_{zv} = i|k_{zv}|$. Hence,

$$I^{(p)}(\omega, k_\rho) = \ln \left(1 - \tilde{R}_{V, [k-1]}^{(p)} \tilde{R}_{V, [k+1]}^{(p)} e^{-2|k_v|l} \right). \quad (21)$$

$I^{(p)}(\omega, k_\rho)$ as defined in Eq. (21) is equally valid in the upper and lower half planes because there is no branch cut in the region $0 < \omega < ck_\rho$. $\tilde{R}_{V, [k-1]}^{(p)}$, $\tilde{R}_{V, [k+1]}^{(p)}$, and $e^{i2k_{zv}l}$ are evaluated using $k_{zv} = |k_{zv}|$ in the upper half plane for $\omega > ck_\rho$. $I^{(p)}(\omega, k_\rho)$ is given by

$$\begin{aligned} I^{(p)}(\omega, k_\rho) &= I_{uhp}^{(p)}(\omega, k_\rho) \\ &= \lim_{l_v \rightarrow \infty} \ln \left[\frac{1 - \tilde{R}_{V, [k-1]}^{(p)} \tilde{R}_{V, [k+1]}^{(p)} e^{i2|k_{zv}|l}}{1 - \tilde{R}_{V, [k-1]}^{(p)} \tilde{R}_{V, [k+1]}^{(p)} e^{i2|k_{zv}|l_v}} \right]. \end{aligned} \quad (22)$$

The subscript *uhp* stands for ‘‘upper half plane.’’ In the lower half plane, $I^{(p)}(\omega, k_\rho)$ for $\omega > ck_\rho$ is evaluated by calculating $\tilde{R}_{V, [k-1]}^{(p)}$ and $\tilde{R}_{V, [k+1]}^{(p)}$ using the definition $k_{zv} = -|k_{zv}|$. By using Eq. (18), $I^{(p)}(\omega, k_\rho)$ in the lower half plane can be related to $I_{lhp}^{(p)}(\omega, k_\rho)$ as

$$\begin{aligned} I^{(p)}(\omega, k_\rho) &= I_{lhp}^{(p)}(\omega, k_\rho) \\ &= \lim_{l_v \rightarrow \infty} \ln \left[\frac{1 - \tilde{R}_{V, [k-1]}^{(p)-1}(|k_{zv}|) \tilde{R}_{V, [k+1]}^{(p)-1}(|k_{zv}|) e^{-i2|k_{zv}|l}}{1 - \tilde{R}_{V, [k-1]}^{(p)-1}(|k_{zv}|) \tilde{R}_{V, [k+1]}^{(p)-1}(|k_{zv}|) e^{-i2|k_{zv}|l_v}} \right] \\ &= I_{uhp}^{(p)}(\omega, k_\rho) + i2|k_{zv}|(l - l_v). \end{aligned} \quad (23)$$

The subscript *lhp* stands for ‘‘lower half plane.’’ The function $I^{(p)}(\omega, k_\rho)$ in the left half plane can be obtained through the following symmetry relation: $I^{(p)}(-|\omega|, k_\rho) = [I^{(p)}(|\omega|, k_\rho)]^*$.

$I^{(p)}(\omega, k_\rho)$ as evaluated along the real frequency axis in the upper and lower half planes are not equal, the difference coming from the $i2|k_{zv}|(l - l_v)$ term in Eq. (23). However, U_v , as we defined in Eq. (17), does not suffer from this term since the $i2|k_{zv}|(l - l_v)$ term is common to both $\lim_{l_v \rightarrow \infty} \ln\left(\frac{D_v^{(p)}(\omega, k_\rho; l)}{D_v^{(p)}(\omega, k_\rho; l_v)}\right)$ and $\lim_{L_v \rightarrow \infty} \ln\left(\frac{D_{\text{SIR}}^{(p)}(\omega, k_\rho; l+l_1+l_2)}{D_{\text{SIR}}^{(p)}(\omega, k_\rho; L_v)}\right)$ and cancel each other.

The singularities of the integrand correspond to poles of $\coth\left[\frac{\hbar\omega}{2k_B T}\right]$ and branch points of the \ln function at zeros of $D_v^{(p)}(\omega, k_\rho; l)$ (modes of the electrodynamic system shown in Fig. 2). Following the arguments of Diaz and Alexopoulos [34], $D_v^{(p)}(\omega, k_\rho; l)$ can have only zeros or pole singularities. We further assume that poles of $D_v^{(p)}(\omega, k_\rho; l)$, if present, are independent of l and hence cancel with the contribution from $D_v^{(p)}(\omega, k_\rho; l_v \rightarrow \infty)$. Because of the analyticity of $D_v^{(p)}(\omega, k_\rho; l)$, it can be written as

$$D_v^{(p)}(\omega, k_\rho; l) = \left(1 - \frac{\omega}{\omega_{m, k_\rho}(l)}\right) \mathcal{D}_m^{(p)}(\omega, k_\rho; l) \quad (24)$$

in a neighborhood of $\omega_{m, k_\rho}(l)$ in which $\mathcal{D}_m^{(p)}(\omega, k_\rho; l)$ is a regular function. Because of the \ln function, a branch cut of the form shown in Fig. 3 is present at each normal mode ω_{m, k_ρ} . The contour path for integrating $\lim_{l_v \rightarrow \infty} \ln\left(\frac{D_v^{(p)}(\omega, k_\rho; l)}{D_v^{(p)}(\omega, k_\rho; l_v)}\right) - \lim_{L_v \rightarrow \infty} \ln\left(\frac{D_{\text{SIR}}^{(p)}(\omega, k_\rho; l+l_1+l_2)}{D_{\text{SIR}}^{(p)}(\omega, k_\rho; L_v)}\right)$ is shown in Fig. 3. Because of Eq. (23), the integrals along $C_{R,L}^+$ and $C_{R,U}^+$ (and $C_{R,U}^-$ and $C_{R,L}^-$) cancel each other. As $|\omega + i\xi| \rightarrow \infty$, $\varepsilon \rightarrow 1$ for all materials and all reflection coefficients vanish. Hence, the integrands along $C_{\infty,U}$ and $C_{\infty,L}$ vanish. Since the integral along the contour in Fig. 3 is zero, the sum of residues at all poles of the integrand within the contour must also equal zero by Cauchy's residue theorem.

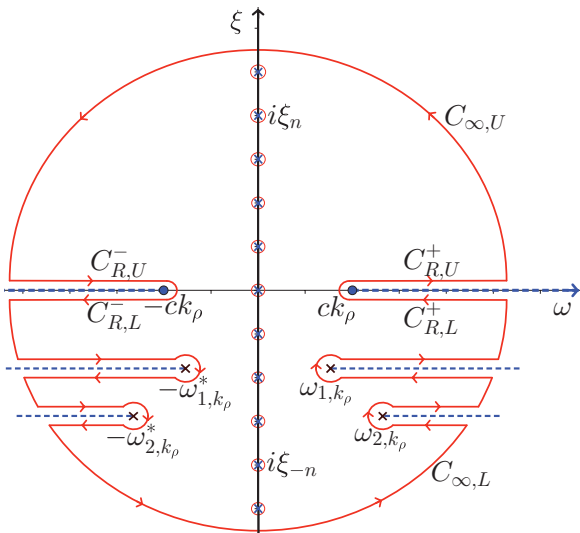


FIG. 3. (Color online) The zeros of $D(\omega; k_\rho)$, are labeled ω_{1, k_ρ} , $\omega_{2, k_\rho}, \dots, \omega_{N, k_\rho}$ and $-\omega_{1, k_\rho}^*$, $-\omega_{2, k_\rho}^*, \dots, -\omega_{N, k_\rho}^*$. The poles on the ξ axis are poles of $\coth(\hbar\omega/2k_B T)$. The red lines are contours for integration.

The contribution from each normal mode at ω_{m, k_ρ} to the contour integral is given by

$$\begin{aligned} & \frac{i\hbar}{4\pi} \int_0^\infty dx (i0 - i2\pi) \left\{ \coth \left[\frac{\hbar(\omega_{m, k_\rho}(l) + x)}{2k_B T} \right] \right. \\ & \quad \left. - \coth \left[\frac{\hbar[\omega_{m, k_\rho}(l_v \rightarrow \infty) + x]}{2k_B T} \right] \right\} \\ & = -k_B T \left\{ \ln \left[2 \sinh \left(\frac{\hbar\omega_{m, k_\rho}(l)}{2k_B T} \right) \right] \right. \\ & \quad \left. - \ln \left[2 \sinh \left(\frac{\hbar\omega_{m, k_\rho}(l_v \rightarrow \infty)}{2k_B T} \right) \right] \right\}. \quad (25) \end{aligned}$$

Clearly, the contribution in Eq. (25) is the change in free energy of a normal mode as the vacuum gap is changed from $l_v \rightarrow \infty$ to $l_v = l$. The $i0$ and $i2\pi$ terms in Eq. (25) are contributions from either side of the branch cut at normal mode frequencies. In deriving Eq. (25), we are assuming that ω_{m, k_ρ} does not coincide with any of the Matsubara frequencies. The contribution from $-\omega_{m, k_\rho}^*$ is the complex conjugate of Eq. (25). The contribution from all the Matsubara frequencies is given by

$$-k_B T \sum_{n=-\infty}^{\infty} \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \sum_{p=e, h} \lim_{l_v \rightarrow \infty} \ln \left[\frac{D_v^{(p)}(i\xi_n, k_\rho; l)}{D_v^{(p)}(i\xi_n, k_\rho; l_v)} \right]. \quad (26)$$

In both Eqs. (25) and (26), the contributions of cavity modes corresponding to the imperfect mirror alone are suppressed in order to make the expression compact. The Lifshitz formula for U_v , corresponding to the contributions from positive Matsubara frequencies, is given by

$$\begin{aligned} U_v(l) & = -k_B T \sum_{n=0}^{\infty} \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \\ & \quad \times \sum_{p=e, h} \lim_{l_v \rightarrow \infty} \ln \left[\frac{D_v^{(p)}(i\xi_n, k_\rho; l)}{D_v^{(p)}(i\xi_n, k_\rho; l_v)} \right]. \quad (27) \end{aligned}$$

From Eqs. (25)–(27), we can relate U_v to the free energies of normal modes as follows:

$$\begin{aligned} U_v(l) & = U_{\text{nor}}(l) + \frac{k_B T}{2} \sum_{n=0}^{\infty} \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \sum_{p=e, h} \lim_{l_v \rightarrow \infty} \\ & \quad \times \ln \left[\frac{D_v^{(p)}(i\xi_{-n}, k_\rho; l) / D_v^{(p)}(i\xi_n, k_\rho; l)}{D_v^{(p)}(i\xi_{-n}, k_\rho; l_v) / D_v^{(p)}(i\xi_n, k_\rho; l_v)} \right], \quad (28) \end{aligned}$$

where $U_{\text{nor}}(l)$ is the sum of free energy of each normal mode and is given by

$$\begin{aligned} U_{\text{nor}}(l) & = k_B T \int_0^\infty \frac{dk_\rho k_\rho}{2\pi} \\ & \quad \times \sum_m \text{Re} \left\{ \lim_{l_v \rightarrow \infty} \ln \left[\frac{\sinh[\hbar\omega_{m, k_\rho}(l)/2k_B T]}{\sinh[\hbar\omega_{m, k_\rho}(l_v)/2k_B T]} \right] \right\}. \quad (29) \end{aligned}$$

In Eq. (29), the \sum_m is performed over modes only in the right half plane. Equations (28) and (29) imply that the van der Waals energy is not only composed of the sum of free energies of normal modes but also contributions from poles at negative Matsubara frequencies. Only when $D_v^{(p)}(i\xi_{-n}, k_\rho; l) =$

$D_v^{(p)}(i\xi_n, k_\rho; l)$, i.e., when $\varepsilon(i\xi_{-n}) = \varepsilon(i\xi_n)$ and $\mu(i\xi_{-n}) = \mu(i\xi_n)$, can the Lifshitz energy be expressed in terms of the sum of free energies of normal modes alone. Only in the high-temperature limit, when the $n = 0$ term dominates the contributions to U_v and U_{nor} , can the van der Waals energy be directly written in terms of the free energy of normal modes since $\varepsilon(i\xi_n), \varepsilon(i\xi_{-n}) \rightarrow 1$ (and similarly for μ) as $2k_B T l / c\hbar \rightarrow \infty$.

IV. DISCUSSION

Keeping in mind that prior works related to the mode-summation method applied to van der Waals interactions are numerous, we first summarize the findings of a representative subset of relevant prior literature [9,15–19,33]. If all materials are assumed to be nondissipative, the zeros of $\tilde{D}_v^{(p)}(\omega + i\xi, k_\rho)$ lie entirely along the real frequency axis. Nesterenko and Pirozhenko [19] consider modifications to the contour used in Ref. [33] because of the presence of poles as well as branch points on the real axis. Bordag [9] uses the low-temperature equivalent of Eq. (1) to analyze the van der Waals energy between two parallel plates made of dispersive but nondissipative materials but takes into account contributions from not only surface modes but also photonic and waveguide modes.

In a series of publications, Intravaia and co-workers [15–18] attempt to address the role of dissipation in determining the van der Waals energy between two metallic, dissipative (described by the Drude model), semi-infinite mirrors by the mode-summation method. They use a “system + bath” approach that has also been used in the quantum mechanical analysis of a damped harmonic oscillator [35,36]. Their main result is that the formula for zero-temperature free energy per mode [low-temperature equivalent of Eq. (1)] has to be modified when the normal mode frequency lies on the negative imaginary frequency axis (they call such modes “eddy current modes”). In our analysis, this corresponds to the case when the mode frequency coincides with any of the negative Matsubara frequencies which lead to a minor modification of Eq. (25). We have not expounded on this particular case since mode frequencies are usually complex and lie on either side of the imaginary frequency axis. Moreover, summation of mode energies is not a practical idea for computing van der Waals energy. The expression that is eventually used for computing van der Waals energy is Eq. (14) or (15), which we have derived without relying on any particular formula for free energy per mode.

The main implication of Eqs. (28) and (29), as recognized by Ninham *et al.* [33], is that the van der Waals energy of a vacuum cavity in a multilayer system can not be expressed simply as a sum of free energies of normal modes of the system, even when correction of the form suggested by Intravaia *et al.* [18] is taken into account, when any of the layers is dissipative. An analogy to this observation can be seen in the quantum mechanical analysis of a damped harmonic oscillator [35,36]. Although not the same quantity as free energy, the position autocorrelation function $J(\tau)$ in Ref. [35], Eqs. (3.1) and (3.2), has similar features to U_v [Eq. (16)]. After evaluating the integral by contour integration [35], Eq. (3.5), $J(\tau)$ can be expressed for $\tau \geq 0$ in terms of contributions from the poles of the susceptibility function (corresponding to U_{nor})

and contributions from negative Matsubara frequencies. The Lifshitz-type form for $J(\tau)$ is shown in Eq. (3.9) of Ref. [35]. Clearly, the mode contribution alone is insufficient to obtain the correct form of the position autocorrelation function.

While we do not have a satisfactory explanation for the fact that mode energies alone are insufficient to capture the entire van der Waals energy, we think it can be done so using the “system + bath + coupling” approach [18,35,36]. The modes of the electromagnetic field in free space is the system under consideration. The introduction of any material other than vacuum immediately introduces the “bath,” which is the collection of harmonic oscillators that represent the material. The dissipation that is included in the dielectric and magnetic response of the material is because of the interaction between the system (electromagnetic modes) and the bath. It can be argued that the temperature of the medium is related to the “bath” alone and neither the system nor the coupling between the two. Although the free energy of normal modes of the electromagnetic field includes the influence of dissipation (as does the complex resonance frequency of a damped harmonic oscillator), this alone, as represented by Eq. (29), may be insufficient to capture the thermodynamics of coupling between electromagnetic fields and matter. While incomplete, we do not have a better answer as to why the free energy of normal modes is insufficient to capture the van der Waals energy of the multilayer system with dissipative materials.

V. SUMMARY

We have derived a method to determine the van der Waals energy and pressure in a dissipative material within a planar multilayer object with arbitrary number of layers. It is shown to be a hybrid of the fluctuational electrodynamics (Lifshitz) method and energy conservation. Like Lifshitz, we use Rytov’s theory of fluctuational electrodynamics and like Casimir (and others) we use principle of energy conservation to extend Lifshitz’ theory to the case of dissipative materials. Unlike Casimir (and others), we do not rely on the assumption that the van der Waals free energy can be computed by adding the free energy of each mode. We have also shown that van der Waals energy and pressure in a dissipative material can be obtained by performing the analysis entirely along the real frequency axis.

We have shown that the integrals [Eqs. (14) and (15)] for van der Waals energy along the real axis can be converted into the form derived by Lifshitz or similar to the summation of free energy of normal modes (but not identical) by pursuing contour integration in the upper and lower halves of the complex frequency plane, respectively. In fact, in computational electrodynamics, it is usual to complete the path of integration in the lower half plane in order to obtain the transient response due to a source ([32], pp. 214 and 215). In view of this, it is remarkable that it was Lifshitz’ idea of completing the path in the upper half plane that has proven to be more robust when analyzing van der Waals interaction in dissipative media. Recent works notwithstanding [18,19], the quest to interpret the expression for van der Waals free energy in terms of the free energy of normal modes of the planar multilayer system within the confines of macroscopic electrodynamics can be

solved only when the question of finite dissipation is addressed in its entirety. To the best of our understanding, nothing new is gained by employing the mode-summation method over the fluctuational electrodynamics method.

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