## Thermal Casimir effect between random layered dielectrics

David S. Dean,<sup>1,2</sup> R. R. Horgan,<sup>1,3</sup> Ali Naji,<sup>1,4</sup> and R. Podgornik<sup>1,5,6,7</sup> <sup>1</sup>Kavli Institute for Theoretical Physics, University of California, Santa Barbara, California 93106, USA

<sup>2</sup>Laboratoire de Physique Théorique, IRSAMC, Universite Paul Sabatier, 118 Route de Narbonne, 31062 Toulouse Cedex 4, France

<sup>3</sup>DAMTP, CMS, University of Cambridge, Cambridge CB3 0WA, United Kingdom

<sup>4</sup>Department of Physics and Department of Chemistry and Biochemistry, University of California, Santa Barbara, California 93106, USA

<sup>5</sup>Department of Physics, Faculty of Mathematics and Physics, University of Ljubljana SI-1000 Ljubljana, Slovenia

<sup>6</sup>Department of Theoretical Physics, J. Stefan Institute, SI-1000 Ljubljana, Slovenia

<sup>7</sup>Laboratory of Physical and Structural Biology, National Institutes of Health, Maryland 20892, USA

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We study the thermal Casimir effect between two thick slabs composed of plane-parallel layers of random dielectric materials interacting across an intervening homogeneous dielectric. It is found that the effective interaction at long distances is self-averaging and is given by a description in terms of effective dielectric functions. The behavior at short distances becomes random (sample dependent) and is dominated by the local values of the dielectric function proximal to each other across the dielectrically homogeneous slab.

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Systems with spatially varying dielectric functions exhibit effective van der Waals interactions arising from the interaction between fluctuating dipoles in the system [1,2]. These fluctuation interactions have two distinct components: (i) a classical or thermal component due to the zero-frequency response of the dipoles and (ii) a quantum component due to the nonzero-frequency or quantum response of the dipoles. Despite the clear physical differences in these contributions, the mathematical computation of the corresponding interaction is almost identical and boils down to the computation of an appropriate functional determinant. The full theory taking into account both of these component interactions is the celebrated Lifshitz theory of van der Waals interactions [3], based on boundary conditions imposed on the electromagnetic field at the bounding surfaces and the fluctuationdissipation theorem for the electromagnetic potential operators. From the Lifshitz theory one can derive the original Casimir interaction [4] by taking the limit of zero temperature and ideally polarizable bounding surfaces. In this respect the Lifshitz theory is nothing but a proper finite temperature and realistic boundary conditions generalization of the Casimir interaction and thus van der Waals interactions are indeed nothing else but the thermal Casimir effect. The major mathematical problems in the computation of Casimir-type interactions (setting aside the experimental and theoretical challenges to determine the correct dielectric behavior) are (i) the application of the Lifshitz approach to nontrivial geometries and (ii) taking into account local inhomogeneities in the dielectric properties of the media, always present in realistic systems. In this paper we will address the latter.

We consider thermal Casimir interaction for the case where the local dielectric function is a random variable in the transverse direction. Specifically we will consider the interaction between two thick parallel dielectric slabs, separated by a homogenous dielectric medium; see Fig. 1. The thickness of both disordered dielectric slabs are  $L_1$  and  $L_2$ , respectively, and their separation is denoted by  $\ell$ . In what follows we will study the limit of infinite slabs, i.e.,  $L_1, L_2 \rightarrow \infty$ . The dielectric response within the two slabs is constant in the planes perpendicular to the slab normal, but varies in the direction of the surface normal. It is well known that this problem can be solved in the case where the dielectric constants of the slabs do not vary [2] and the result can be tentatively applied to the case of fluctuating dielectric functions via an effective-medium theory which consists of replacing the fluctuating dielectric functions by an effective (spatially constant within each of the slabs) dielectric tensor. The most commonly used approximation is that, where the local dielectric tensor is replaced by the effective dielectric tensor [1,2], i.e.,

$$\boldsymbol{\epsilon}_{ij}(\mathbf{x}) \to \boldsymbol{\epsilon}_{ij}^{(e)}, \tag{1}$$

where the bulk dielectric tensor is defined via  $\epsilon_{ii}^{(e)} \langle E_i \rangle$  $=\langle \epsilon_{ij} E_{j} \rangle$ . The use of the effective dielectric function is not easily justifiable mathematically as an approximation, although physically the effective dielectric function clearly



FIG. 1. (Color online) A schematic presentation of the model. Two finite slabs, (1) and (2), with disordered plane-parallel dielectric layers interacting across a dielectrically homogeneous slab of thickness  $\ell$ . z axis is perpendicular to the plane of the slabs.

does capture the bulk response to constant electric fields. We shall see that, for the random layered dielectric model studied here, the effective dielectric constant approximation of Eq. (1) does in fact give the correct value of the thermal Casimir interaction when the two slabs are widely separated. This can be expected on physical grounds as the fluctuating electromagnetic field modes with smallest wave vectors (corresponding to variations on large scales) dominate the Casimir interaction for large interslab separation. The dielectric response of the material to a constant electric field is given by the effective dielectric constant and if the wave-vector dependent response is suitably analytic near  $\mathbf{k}=\mathbf{0}$  we expect that  $\boldsymbol{\epsilon}_{ie}^{(e)}(\mathbf{k}) \sim \boldsymbol{\epsilon}_{ie}^{(e)}$  for  $|\mathbf{k}| \leq 1$ .

In this paper we introduce a path-integral formalism to compute the thermal Casimir free energy between two semiinfinite dielectric slabs which are composed of layers with varying dielectric function. Our formulation allows us to show rigorously that for large interslab separations the leading-order contribution to the interaction is self-averaging and is equivalent to that obtained by replacing each slab with a homogeneous (though nonisotropic medium) with a dielectric tensor equal to the effective (bulk) dielectric tensor of the disordered medium. The short-distance behavior of the interaction is random and we show, as would be expected on physical grounds, that it is dominated by the precise value of the dielectric constants at the two opposing slab faces.

The Hamiltonian for the zero-frequency fluctuations of the electrostatic field in a dielectric medium is given by the classical electromagnetic field energy

$$H[\phi] = \frac{1}{2} \int d\mathbf{x} \,\epsilon(\mathbf{x}) [\nabla \phi(\mathbf{x})]^2, \qquad (2)$$

and the corresponding partition function is given by the functional integral  $Z = \int d[\phi] \exp(-\beta H[\phi])$ . Differences in dielectric functions lead to the thermal Casimir effect. Here we will consider layered systems where the dielectric function  $\epsilon$ depends only on the *z* direction  $\epsilon(\mathbf{x}) = \epsilon(z)$ . If we express the field  $\phi$  in terms of its Fourier modes in the plane perpendicular to *z*, and we take the area perpendicular to *z* as *A*, with wave vector  $\mathbf{k} = (k_x, k_y)$ , then the Hamiltonian can be written as  $H = \sum_{\mathbf{k}} H_{\mathbf{k}}$  with

$$H_{\mathbf{k}} = \frac{1}{2} \int dz \, \boldsymbol{\epsilon}(z) \Biggl( \left| \frac{d \tilde{\phi}(z, \mathbf{k})}{dz} \right|^2 + \mathbf{k}^2 |\tilde{\phi}(z, \mathbf{k})|^2 \Biggr).$$
(3)

Therefore the partition function can be expressed as a sum over the partition functions of individual modes  $Z_k$  as  $\ln(Z) = \sum_k \ln(Z_k)$  where

$$Z_{\mathbf{k}} = \int d[X] \exp\left(-\frac{1}{2} \int dz \,\epsilon(z) \left[\left(\frac{dX}{dz}\right)^2 + k^2 X^2\right]\right). \tag{4}$$

Here  $k = |\mathbf{k}|$  and we have taken into account that the field  $\phi$  is real.

The problem of computing the interaction between slabs composed of layers of finite thickness can be studied using a transfer-matrix-like method [5]. However we will use a method based on the Feynman path integral instead, which is particularly well suited to the study of systems where the dielectric function can vary continuously [6]. If we specify the starting and finishing points of the above path integral, we see that it has to be of a harmonic-oscillator form defined by

$$K(x,y;z',z) = \int_{X(z')=x}^{X(z)=y} d[X] \exp\left\{-\frac{1}{2}\int dz M(z) \left[\left(\frac{dX}{dz}\right)^2 + \omega^2 X^2\right]\right\},$$
(5)

which can be computed using the generalized Pauli–van Vleck formula [6,7] telling us that *K* must have the general form

$$K(x,y;z',z) = \left[\frac{b(z',z)}{2\pi}\right]^{1/2} \exp\left[-\frac{1}{2}a_i(z',z)x^2 - \frac{1}{2}a_f(z',z)y^2 + b(z',z)xy\right].$$
(6)

We may now write down an evolution equation for the coefficients  $a_i$ ,  $a_f$ , and b using the Markovian property of the path integral (in fact this is how one can prove the generalized Pauli–van Vleck formula:  $K(x,y;z',z+\zeta) = \int dw K(x,w;z',z)K(w,y;z,z+\zeta)$  [6,7]) to find the following evolution equations for  $a_i$ ,  $a_f$ , and b:

$$\frac{\partial a_i(z',z)}{\partial z} = -\frac{b^2(z',z)}{M(z)}, \quad \frac{\partial b(z',z)}{\partial z} = -\frac{b(z',z)a_f(z',z)}{M(z)},$$
$$\frac{\partial a_f(z',z)}{\partial z} = M(z)\omega^2(z) - \frac{a_f^2(z',z)}{M(z)}.$$
(7)

We thus find that the  $\ell$ -dependent part of the free energy of the mode **k** (up to a bulk term which can be subtracted off to get the effective interaction) is given by

$$F_{\mathbf{k}} = \frac{k_B T}{2} \ln \left\{ 1 - \frac{[a_f^{(1)}(k) - \epsilon_0 k] [a_f^{(2)}(k) - \epsilon_0 k]}{[a_f^{(1)}(k) + \epsilon_0 k] [a_f^{(2)}(k) + \epsilon_0 k]} e^{-2k\ell} \right\}, \quad (8)$$

and the total  $\ell$  dependent free energy is  $F = \sum_{k} F_{k}$ . Here  $a_{f}^{(1,2)}(k)$  are the solutions to Eq. (7) evaluated at the opposing faces of each slab [(1) and (2).

In order to evaluate the integrals of  $a_f^{(1,2)}(k)$ , one first has to solve equations of motion Eq. (7) to get the *z* dependence of  $a_f(k,z)$  and then proceed to the integrals that enter Eq. (8). The evolution equation for  $a_f(k)$  for either slab can be read off from Eq. (7) and is given by

$$\frac{da_f(k,z)}{dz} = \epsilon(z)k^2 - \frac{a_f^2}{\epsilon(z)},\tag{9}$$

where we have dropped the explicit dependence on  $z'=-L_1$ . An appropriate Hopf-Cole transformation [7] shows this formalism to be equivalent to the transfer-matrix method [5] or to the density-functional method [8] for evaluating the van der Waals forces. This nonlinear formulation of an essentially linear problem in fact simplifies the analysis of the effect of disorder in a similar way as it does in quantum problems [9]. If we now write  $a_f^{(i)}(k,z) = k\alpha^{(i)}(k,z)$  and if the distributions of the  $\alpha^{(i)}(k,z) = y$  are given by  $p_i(k,y)$  then we find that, in three dimensions the average of the  $\ell$  dependent free energy is given by

$$\langle F \rangle = \frac{k_B T A}{4\pi} \int dkk \int dy_1 \int dy_2 p_1(k, y_1) p_2(k, y_2)$$
$$\times \ln \left( 1 - \frac{(y_1 - \epsilon_0)(y_2 - \epsilon_0)}{(y_1 + \epsilon_0)(y_2 + \epsilon_0)} e^{-2k\ell} \right), \tag{10}$$

where the angled bracket on the lhs indicates the disorder average over the dielectric function within the slabs and we have assumed that the realizations of the disorder in the two slabs are independent.

Let us first investigate the form of van der Waals interaction free energy in the limit of large separations between the two slabs. The equation obeyed by  $\alpha$  can be written as

$$\frac{d\alpha(k,\zeta)}{d\zeta} = \epsilon(\zeta/k) - \frac{\alpha^2}{\epsilon(\zeta/k)},\tag{11}$$

with  $\zeta = zk$ . When k is small  $\epsilon(\zeta/k)$  varies very rapidly and thus becomes decorrelated from the value of  $\alpha$ . The Laplace transform for the probability density function of  $\alpha$  is defined by  $\tilde{p}(k,s,\zeta) = \int_0^\infty dy \exp(-sy)p(k,y,\zeta) = \langle \exp[-s\alpha(k,\zeta)] \rangle$ , and, from the equation of motion Eq. (11), obeys

$$-\frac{1}{s}\frac{d\tilde{p}(k,s,\zeta)}{d\zeta} = \left\langle \epsilon(\zeta/k)\exp[-s\alpha(k,\zeta)] - \frac{\alpha^2}{\epsilon(\zeta/k)}\exp[-s\alpha(k,\zeta)] \right\rangle.$$
(12)

Assuming that k is small and thus  $\alpha(k,\zeta)$  and  $\epsilon(\zeta/k)$  are decorrelated we can write

$$-\frac{1}{s}\frac{d\tilde{p}(k,s,\zeta)}{d\zeta} = \langle \epsilon \rangle \tilde{p}(k,s,\zeta) - \langle 1/\epsilon \rangle \frac{d^2}{ds^2} \tilde{p}(k,s,\zeta).$$
(13)

As we are interested in the limit of thick slabs it suffices to know the equilibrium distribution of this equation which is given by  $\lim_{\zeta \to \infty} \tilde{p}(k, s, \zeta) = \exp(-\epsilon^* s)$  with

$$\boldsymbol{\epsilon}^* = \sqrt{\frac{\langle \boldsymbol{\epsilon} \rangle}{\langle 1/\boldsymbol{\epsilon} \rangle}}.$$
 (14)

Inverting the Laplace transform then gives the equilibrium distribution  $p(y,k) = \delta(y - \epsilon^*)$  at small k. When  $\ell$  is large the integral in Eq. (10) is dominated by the small k behavior, and we may use the analysis presented above to give the following asymptotic form for the interaction free energy,

$$\langle F \rangle (\ell \to \infty) \sim \frac{k_B T A}{16 \pi \ell^2} \int u du \ln(1 - \Delta_1^* \Delta_2^* e^{-u}) = -\frac{H^* A}{\ell^2},$$
(15)

with  $\Delta_i^* = (\epsilon_i^* - \epsilon_0)/(\epsilon_i^* + \epsilon_0)$  and where  $\epsilon_i^*$  are defined via Eq. (14). The subscript *i* on the angled brackets signifies that we are averaging the dielectric function in the slab *i*. The term  $H^*$  defines an effective disorder-dependent Hamaker coefficient. This therefore justifies physical arguments replacing the random layered material by an effective anisotropic medium where the dielectric tensor is has the form  $\epsilon_{zz}^{(e)} = \epsilon_{\parallel}$  and  $\epsilon_{xx}^{(e)} = \epsilon_{yy}^{(e)} = \epsilon_{\perp}$ , all other terms being zero by symmetry. The

term  $\epsilon_{\parallel}$  is the effective dielectric function in the *z* direction  $\epsilon_{\parallel}^{(e)} = 1/\langle 1/\epsilon \rangle$  and the perpendicular components are given by  $\epsilon_{\perp}^{(e)} = \langle \epsilon \rangle$ . The expressions for  $\epsilon_{\parallel}^{(e)}$  and  $\epsilon_{\perp}^{(e)}$  follow simply from the fact that in the perpendicular direction the dielectric function is obtained by analogy to capacitors in series and in the parallel direction by analogy to capacitors in parallel arrangement [10]. The effective value of  $\epsilon^*$  for this system coincides with that of Eq. (14) above [7]. This result shows that for large separations (where  $\ell$  is much larger than the correlation length of the dielectric disorder) the thermal Casimir interaction free energy is self-averaging and agrees with that given by physical reasoning.

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One would imagine that as the distance between the slabs is reduced, the result will be increasingly dominated by the slab composition at the two opposite faces [2]. Indeed in the small  $\ell$  limit Eq. (10) is dominated by the large *k* behavior. The asymptotic behavior can be extracted if one assumes the *ansatz*,

$$\alpha(z,k) = \sum_{n=0}^{\infty} \frac{\alpha_n(z)}{k^n}.$$
 (16)

Substituting this into Eq. (11) gives the following chain of equations for  $\alpha_n(z)$ :

$$\frac{1}{k}\sum_{n=0}^{\infty}\frac{1}{k^n}\frac{d\alpha_n(z)}{dz} = \epsilon(z) - \frac{1}{\epsilon(z)}\sum_{n,m=0}^{\infty}\frac{\alpha_n(z)\alpha_m(z)}{k^{m+n}}.$$
 (17)

From here it is easy to see that to order O(1) the leading asymptotic result of Eq. (22) is given by

$$\alpha_0(z) = \epsilon(z). \tag{18}$$

The equation for the corrections  $(n \ge 1)$  to this asymptotic limit is

$$\frac{d\alpha_{n-1}(z)}{dz} = -\frac{1}{\epsilon(z)} \sum_{m=0}^{n} \alpha_m(z) \alpha_{n-m}(z), \qquad (19)$$

and the next two terms from this expansion yield

$$\alpha_1(z) = -\frac{1}{2} \frac{d\epsilon(z)}{dz},$$
(20)

$$\alpha_2(z) = \frac{1}{4} \frac{d^2 \epsilon(z)}{dz^2} - \frac{1}{8\epsilon(z)} \left(\frac{d\epsilon(z)}{dz}\right)^2.$$
 (21)

It is straightforward to realize that these terms generate  $O(1/\ell)$  corrections to the asymptotic result which are subdominant when  $\ell$  is large. Thus to the leading order,

$$\alpha(z,k) \approx \alpha_0(z) = \epsilon(z) \tag{22}$$

and from here it follows straightforwardly that

$$\lim_{k \to \infty} p_i(y,k) = \rho_i(\epsilon), \qquad (23)$$

where  $\rho_i$  is the probability density function of  $\epsilon(z)$  in medium *i*. This result is easily understood from the physical discussion above. The average of the thermal Casimir interaction free energy Eq. (10) in the small separation limit is thus given by

$$\langle F \rangle (\ell \to 0) \sim \frac{k_B T A}{16 \pi \ell^2} \int u du \int \rho_1(\epsilon_1) \rho_2(\epsilon_2) d\epsilon_1 d\epsilon_2 \times \ln(1 - \Delta_1 \Delta_2 e^{-u}),$$
 (24)

with  $\Delta_i = (\epsilon_i - \epsilon_0)/(\epsilon_i + \epsilon_0)$ . The forms of the thermal Casimir interaction free energy are thus given by Eqs. (15) and (24) in the small and large interslab separation limits, respectively.

We have thus obtained the limiting behavior of the thermal Casimir effect in the limit of large separation between the slabs, where the free energy is given by self-averaging and thus the distributions of  $\alpha(k,z)$  are strongly peaked. It can be shown [7] that the large separation attraction between two (statistically identical) homogeneous media (with  $\epsilon = \langle \epsilon \rangle$ ) is stronger than that between the two fluctuating media if  $\langle 1/\epsilon \rangle^{-1} > \epsilon_0$ . However it is always weaker if  $\langle \epsilon \rangle < \epsilon_0$ . We thus see that, depending on the details of the distribution of the fluctuating dielectric response in the two slabs and the dielectric response of the medium in between, the effective interaction at large interslab separations can be stronger or weaker than that for a uniform medium with a dielectric constant equal to the mean dielectric function of the fluctuating media.

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For small separations the interaction free energy is a random variable and its mean value is obtained by averaging over the probability density functions of the dielectric functions of the media composing the opposing surfaces of the interacting slabs. The intermediate length scales can be analyzed via perturbation theory [7] and possibly there exist models of disorder that can be treated exactly. The nonlinear formulation of the problem presented here should be equally useful to treat the case of deterministically varying dielectric functions and could open up a useful computational framework for designing materials where the effective interaction can be tuned, to induce attractive or repulsive forces depending on the separation, for practical applications [11].

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