Casimir dispersion forces and orientational pairwise additivity

Ramin Golestanian
Institute for Theoretical Physics, University of California, Santa Barbara, California 93106
and Institute for Advanced Studies in Basic Sciences, Zanjan 45195-159, Iran
(Received 28 February 2000)

A path-integral formulation is used to study the fluctuation-induced interactions between manifolds of arbitrary shape at large separations. It is shown that the form of the interactions crucially depends on the choice of the boundary condition. In particular, whether or not the Casimir interaction is pairwise additive is shown to depend on whether the ''metallic'' boundary condition corresponds to a ''grounded'' or an ''isolated'' manifold.

PACS number(s): 82.70.Dd, 05.70.Jk, 05.70.Np

I. INTRODUCTION AND SUMMARY

External objects that are immersed in a fluctuating medium, and modify the fluctuations in their vicinity, experience induced interactions with one another [1–5]. These interactions are most often independent of the structural details, and are in turn highly sensitive to the geometry of the objects and their mutual arrangements while immersed in the medium.

The strong dependence of these interactions on the shape of the objects raises the issue of *pairwise additivity*: Is it possible to express the fluctuation–induced interaction between two extended bodies as the sum of a pair potential, or the interaction between several bodies as the sum of two-body interactions?

It is well known that a pairwise summation of the van der Waals interaction gives the correct power law for the Casimir energy [3]. Let us take a pair potential of the form $-A/r^n$, with $n=6$ for the thermal case and $n=7$ for the quantum case [2], and A being a constant to be determined. If one tries to fix the coefficient by summing the pair potential over two bodies and equating the result to the expression for the Casimir interaction between the bodies, one finds out that a different coefficient is needed for every geometry.

To understand this, one should note that the van der Waals interaction is due to dipolar fluctuations. When two extended bodies are at a close separation, one can show that the fluctuations of all the multipoles in fact contribute comparably to the Casimir energy, and thus summation of the contribution due to the dipolar fluctuations cannot by itself account for the interaction [6]. When the bodies are at large separations (larger than their typical sizes), the contribution due to higher multipoles is in fact systematically weaker. However, there is still a discrepancy between the sum of a (van der Waals) pair potential, and the contribution of the dipolar fluctuations to the Casimir energy. In the spirit of a (second order) perturbation theory, the correct way of calculating the dipolar Casimir energy is to consider the pairwise sum of the dipole-dipole interactions over the two bodies, and then square it and take the average. This is clearly in contrast to the pairwise summation of the van der Waals interaction, which corresponds to taking the square of the local dipolar fluctuations and averaging, and then summing over the two bodies.

The same picture can help us answer the second question. Many-body interactions can be expressed as the sum of many-body interactions of the multipoles of different bodies in the medium. When extended bodies are at close separations, and all the multipoles have comparable contributions, many-body interactions of nontrivial forms result [7–9]. On the other hand, for bodies at large separations, the leading-order contribution comes from the sum of two-body interactions of the lowest nonvanishing multipole [10].

In this paper, we study the issue of orientational pairwise additivity [10,11], which is to determine whether the orientational dependence of the interactions could be obtained from the summation of a pair potential. A path-integral formulation is used to study the fluctuation-induced interactions between manifolds of arbitrary shape at large separations, in the context of a multipole expansion. It is shown that the form of the interaction crucially depends on whether the manifolds are *grounded* or *isolated* in an electrostatic analogy. In the grounded case, the manifolds are connected to a *charge reservoir* to maintain a constant *potential*, and thus the leading fluctuations are *monopolar*. Isolated manifolds, however, are constrained to have fixed overall charges, and can only undergo *dipolar* fluctuations. The leading interaction between grounded manifolds is found to be of the form $[(\text{monopole})\text{-}(\text{monopole})]^2$, and is independent of their shapes and orientations. The leading shape dependent term comes from the $[(\text{monopole})\text{-}(\text{dipole})]^2$ term, which gives rise to orientational dependencies that are pairwise additive. The interaction between isolated manifolds, however, is dominated by the $[(\text{dipole})\text{-}(\text{dipole})]^2$ term to the leading order, which is *not* pairwise additive.

The rest of the paper is organized as follows. In Sec. II, the path-integral formulation is developed and general expressions are derived for the fluctuation-induced interactions for different types of boundary conditions. In Sec. III, the interactions are examined for the specific examples of symmetric objects such as spheres, and also highly asymmetric objects such as rods and disks, where the above features can be manifestly understood. Critical fluids are examined in Sec. IV, as a special case, and a conclusion follows in Sec. V.

II. PATH-INTEGRAL FORMULATION

Consider a d-dimensional medium, in which a field ϕ is undergoing thermal fluctuations, and n immersed external

bodies (manifolds) denoted by $M_\alpha (\alpha=1,\ldots,n)$, which modify the fluctuations. Let us assume that the fluctuations are scale-free (massless), and thus can be described by the Hamiltonian

$$\mathcal{H}[\phi]=\frac{K}{2}\int d^d\mathbf{x}(\nabla\phi)^2. \tag{1}$$

The field could represent a component of the electromagnetic field (e.g. the electric potential) in a dielectric medium or vacuum [3], the electrostatic potential in charged fluids at very low salt concentrations [5,12,13], an order-parameter field for a critical binary mixture or a magnetic system [14], a massless Goldstone mode arising from a continuous symmetry breaking [7], or an elastic deformation field for fluctuating membranes and surfaces [10,15,16].

In an electrostatic terminology, which we take up in what follows for simplicity, one can view each manifold as a *conductor* that requires a constant value for the potential field in the whole volume that it encloses. A restricted partition function, which requires a value of ϕ_α for the potential field on the αth manifold, can then be written as

$$\mathcal{Z}[\phi_\alpha]=\int \mathcal{D}\phi(\mathbf{x})\prod_{\alpha=1}^{n}\delta\{\phi|_{M_\alpha}-\phi_\alpha\}e^{-\mathcal{H}[\phi]}. \tag{2}$$

Following Ref. [7], the functional delta functions can next be represented by introducing the Lagrange multiplier fields $\rho_\alpha(\mathbf{x})$ as

$$\begin{aligned}\mathcal{Z}[\phi_\alpha]&=\int \mathcal{D}\phi(\mathbf{x})\prod_{\alpha=1}^{n}\int_{M_\alpha}\mathcal{D}\rho_\alpha(\mathbf{x})\exp\Bigg\{-\frac{K}{2}\int d^d\mathbf{x}(\nabla\phi)^2\\&\quad+i\sum_\alpha\int d^d\mathbf{x}\rho_\alpha(\mathbf{x})[\phi(\mathbf{x})-\phi_\alpha]\Bigg\}\\&=\mathcal{Z}_0\times\prod_{\alpha=1}^{n}\int_{M_\alpha}\mathcal{D}\rho_\alpha(\mathbf{x})\exp\Bigg\{-\frac{1}{2K}\\&\quad\times\sum_{\alpha,\beta}\int d^d\mathbf{x}d^d\mathbf{x}'\rho_\alpha(\mathbf{x})G(\mathbf{x}-\mathbf{x}')\rho_\beta(\mathbf{x}')\\&\quad-i\sum_\alpha\phi_\alpha\int d^d\mathbf{x}\rho_\alpha(\mathbf{x})\Bigg\},\end{aligned} \tag{3}$$

in which

$$G(\mathbf{x}-\mathbf{x}')=(-\nabla^2)^{-1}_{\mathbf{x},\mathbf{x}'}=\frac{1}{S_d(d-2)|\mathbf{x}-\mathbf{x}'|^{d-2}}, \tag{4}$$

with $S_d=2\pi^{d/2}/\Gamma(d/2)$ (the surface area of the d-dimensional sphere), $\mathcal{Z}_0$ is the free partition function, and $\int_{M_\alpha}\mathcal{D}\rho_\alpha(\mathbf{x})$ implies a functional integration only in the region enclosed by M_α. [In other words, the Lagrange multiplier field $\rho_\alpha(\mathbf{x})$ is nonzero only within the volume of M_α.] Note that one should view the $\rho_\alpha(\mathbf{x})$ fields as fluctuating charge-density fields, and Eq.(3) as the partition function of a set of interacting Coulomb plasmas, in the electrostatic context [15].

The fluctuation-induced interactions between the conductors can now be inferred from the above partition function. However, it is important to specify the boundary conditions for the conductors. One possibility is that the conductors are grounded, that is to say they are maintained at a constant fixed potential (Dirichlet boundary condition) by being in contact with a large reservoir of charges; a so-called ''ground.'' In this case, the free energy of the system is obtained as

$$F_{\rm gr}=-k_{\rm B}T\ln\mathcal{Z}[\phi_\alpha=0]. \tag{5}$$

The other possibility is that the conductors are made isolated, and maintain constant amounts of net charges, which we assume to be zero. In this case, the potential field at the conductors can take any value to help maintain the neutrality, and thus the free energy is obtained as

$$F_{\rm is}=-k_{\rm B}T\ln\left(\int_{-\infty}^{+\infty}\prod_\alpha d\phi_\alpha\mathcal{Z}[\phi_\alpha]\right). \tag{6}$$

Note that an isolated conductor has a fixed *overall* charge, which should be contrasted with the case of a Neumann boundary condition where the local surface charge-density $\partial_n\phi$ is fixed.

To further proceed, we focus on the situation in which the manifolds are far from each other, namely, they are at separations much larger than their typical sizes. In this case, we can perform a multipole expansion for the charge-density distribution. For example, the Coulomb interaction between the αth and the βth conductors can be written as ($\alpha\neq\beta$)

$$\begin{aligned}h_{\alpha\beta}&=\int d^d\mathbf{x}d^d\mathbf{x}'\frac{\rho_\alpha(\mathbf{x})\rho_\beta(\mathbf{x}')}{S_d(d-2)|\mathbf{x}-\mathbf{x}'+\mathbf{R}_{\alpha\beta}|^{d-2}}\\&=\frac{Q_\alpha Q_\beta}{S_d(d-2)R_{\alpha\beta}^{d-2}}-\left(\frac{Q_\beta\mathbf{P}_\alpha\cdot\hat{\mathbf{R}}_{\alpha\beta}-Q_\alpha\mathbf{P}_\beta\cdot\hat{\mathbf{R}}_{\alpha\beta}}{S_dR_{\alpha\beta}^{d-1}}\right)\\&\quad+\left(\frac{\mathbf{P}_\alpha\cdot\mathbf{P}_\beta-d\mathbf{P}_\alpha\cdot\hat{\mathbf{R}}_{\alpha\beta}\mathbf{P}_\beta\cdot\hat{\mathbf{R}}_{\alpha\beta}}{S_dR_{\alpha\beta}^{d}}\right)+\ldots,\end{aligned} \tag{7}$$

in which $R_{\alpha\beta}$ is the distance between the two conductors, and the multipoles are defined as

$$Q_\alpha=\int d^d\mathbf{x}\rho_\alpha(\mathbf{x})=\tilde{\rho}_\alpha(\mathbf{k})|_{\mathbf{k}=\mathbf{0}}, \tag{8}$$

$$P_{\alpha,i}=\int d^d\mathbf{x}x_i\rho_\alpha(\mathbf{x})=\frac{1}{i}\frac{\partial}{\partial k_i}\tilde{\rho}_\alpha(\mathbf{k})|_{\mathbf{k}=\mathbf{0}}, \tag{9}$$

with

$$\tilde{\rho}_\alpha(\mathbf{k})=\int d^d\mathbf{x}\rho_\alpha(\mathbf{x})e^{i\mathbf{k}\cdot\mathbf{x}}. \tag{10}$$

We also need to make a similar multipole expansion for the self energy terms at each manifold

$$h_{\alpha\alpha}=\int d^d\mathbf{x}d^d\mathbf{x}'\frac{\rho_\alpha(\mathbf{x})\rho_\alpha(\mathbf{x}')}{S_d(d-2)|\mathbf{x}-\mathbf{x}'|^{d-2}}$$
$$=\int\frac{d^d\mathbf{k}}{(2\pi)^d}\frac{1}{k^2}\tilde{\rho}_\alpha(\mathbf{k})\tilde{\rho}_\alpha(-\mathbf{k}). \quad (11)$$

We can introduce the multipoles, using the Taylor expansion of the charge density in Fourier space

$$\tilde{\rho}_\alpha(\mathbf{k})=\tilde{\rho}_\alpha(\mathbf{0})+\frac{1}{i}\left.\frac{\partial\tilde{\rho}_\alpha(\mathbf{k})}{\partial k_i}\right|_{\mathbf{k}=\mathbf{0}}ik_i+\cdots$$
$$=Q_\alpha+P_{\alpha,i}ik_i+\cdots. \quad (12)$$

The above expansion can be formally viewed as an expansion in powers of kL_α, where L_α is a typical size of the manifold. The expansion is thus convergent only for sufficiently small values of k, corresponding to length scales larger than the size of the manifolds. Since the self energy integral in Eq. (11) involves contributions from higher wave vectors, a multipole expansion for the self energy will be divergent. However, the expansion in Eq. (12) indicates that all the information concerning the first few multipoles of the charge distribution is already contained in the low k behavior of the function $\tilde{\rho}_\alpha(\mathbf{k})$. Since we are only interested in the dependence of the partition function Eq. (3) on the distances $R_{\alpha\beta}$, all we need to know about the self energy is its dependence on the first few multipoles, which is in fact well behaved.

Let us denote the domain of convergence for the expansion in Eq. (12) in k space by $\mathcal{D}_\alpha$. This domain contains the origin, and its shape is determined by the geometry of the conductor. Loosely speaking, its size in each direction is set by the inverse of the size of the conductor in that direction. Now we can restrict the k integral in the self energy only to this domain, and neglect the contribution from the outside of $\mathcal{D}_\alpha$, because all the dependence on the first few multipoles is included in the domain $\mathcal{D}_\alpha$. We thus have

$$h_{\alpha\alpha}=\int_{\mathcal{D}_\alpha}\frac{d^d\mathbf{k}}{(2\pi)^d}\frac{1}{k^2}\tilde{\rho}_\alpha(\mathbf{k})\tilde{\rho}_\alpha(-\mathbf{k})+\cdots$$
$$=\gamma_\alpha Q_\alpha^2+\gamma_{\alpha,ij}P_{\alpha,i}P_{\alpha,j}+\cdots, \quad (13)$$

in which

$$\gamma_\alpha=\int_{\mathcal{D}_\alpha}\frac{d^d\mathbf{k}}{(2\pi)^d}\frac{1}{k^2}, \quad (14)$$

$$\gamma_{\alpha,ij}=\int_{\mathcal{D}_\alpha}\frac{d^d\mathbf{k}}{(2\pi)^d}\frac{k_ik_j}{k^2}, \quad (15)$$

and so forth.

Putting all the pieces together, the $R_{\alpha\beta}$-dependent part of the partition function can be written as

$$\mathcal{Z}[\phi_\alpha]=\int\prod_\alpha dQ_\alpha d\mathbf{P}_\alpha\cdots\exp\left(-i\sum_\alpha\phi_\alpha Q_\alpha\right)$$
$$\times\exp\left\{-\frac{1}{2K}\sum_\alpha[\gamma_\alpha Q_\alpha^2+\gamma_{\alpha,ij}P_{\alpha,i}P_{\alpha,j}+\cdots]\right.$$
$$-\frac{1}{2K}\sum_{\alpha\neq\beta}\left[\frac{Q_\alpha Q_\beta}{S_d(d-2)R_{\alpha\beta}^{d-2}}\right.$$
$$-\frac{\hat{R}_{\alpha\beta,i}(Q_\beta P_{\alpha,i}-Q_\alpha P_{\beta,i})}{S_dR_{\alpha\beta}^{d-1}}$$
$$\left.\left.+\frac{(\delta_{ij}-d\hat{R}_{\alpha\beta,i}\hat{R}_{\alpha\beta,j})P_{\alpha,i}P_{\beta,j}}{S_dR_{\alpha\beta}^d}+\cdots\right]\right\}. \quad (16)$$

Note that we have neglected a Jacobian in changing the measure of integration. However, since the transformation from the charge-density distribution to the multipole description is linear, one can show that the Jacobian is just an uninteresting constant.

Finally, using Eqs. (5) and (16), the interaction free energy for grounded manifolds can be obtained as

$$F_{\rm gr}=-\frac{k_BT}{4S_d^2}\sum_{\alpha\neq\beta}\left[\frac{\gamma_\alpha^{-1}\gamma_\beta^{-1}}{(d-2)^2R_{\alpha\beta}^{2(d-2)}}\right.$$
$$\left.+\frac{(\gamma_\alpha^{-1}\gamma_{\beta,ij}^{-1}+\gamma_{\alpha,ij}^{-1}\gamma_\beta^{-1})\hat{R}_{\alpha\beta,i}\hat{R}_{\alpha\beta,j}}{R_{\alpha\beta}^{2(d-1)}}\right]+O(1/R^{2d}). \quad (17)$$

The first term in Eq. (17) is a squared monopole-monopole interaction, and is independent of the relative orientations of the conductors in space. The second term, on the other hand, has the form of a squared monopole-dipole interaction, and does depend on the orientations through an effective dipole-dipole interaction, which is pairwise additive.

Similarly, for isolated manifolds, Eqs. (6) and (16) yield the interaction as

$$F_{\rm is}=-\frac{k_BT}{4S_d^2}\sum_{\alpha\neq\beta}\frac{\gamma_{\alpha,ik}^{-1}\gamma_{\beta,jl}^{-1}}{R_{\alpha\beta}^{2d}}(\delta_{ij}-d\hat{R}_{\alpha\beta,i}\hat{R}_{\alpha\beta,j})$$
$$\times(\delta_{kl}-d\hat{R}_{\alpha\beta,k}\hat{R}_{\alpha\beta,l})+O(1/R^{2d+2}). \quad (18)$$

Note that the leading term in Eq. (18) is a squared dipole-dipole interaction, and thus it is not orientationally pairwise additive.

III. APPLICATION TO SPECIFIC GEOMETRIES

The multipole expansion allowed us to calculate the general forms of the fluctuation-induced interactions between manifolds of arbitrary shape and with arbitrary orientations with respect to one another, for the two cases of isolated and grounded boundary conditions. All the specific information about the shapes and the orientations of the manifolds are encoded in the γ tensors defined above. This information is in fact of three kinds: (i) the overall magnitude of the tensors

that are set by the typical sizes of the manifolds, (ii) the orientational dependencies that make up the tensorial structure, and are dictated by the structure of the symmetry axes or ''the principal axes'' of the manifolds, and (iii) overall numerical prefactors of order unity. In this section, we try to use symmetry arguments to determine the γ tensors for some simple geometries within the numerical prefactors, without actually specifying the exact shape of the integration domain $\mathcal{D}$. The final piece of information, which is the numerical prefactor, appears to be very sensitive to the exact geometry of the manifold (and thus to that of $\mathcal{D}$), and can in general be calculated using the techniques developed in Ref. [10].

A. Two spheres

The γ tensors for a sphere of radius L can be easily estimated using symmetry: $\gamma_s \sim \int_0^{1/L} k^{d-1} dk/k^2 \sim 1/L^{d-2}$ and $\gamma_{s,ij} \sim \delta_{ij} \int_0^{1/L} k^{d-1} dk \sim \delta_{ij}/L^d$. Using Eqs. (17) and (18), the interaction between a sphere of radius L_1 and another sphere of radius L_2 that is at a distance R reads

$$F_{\rm gr}^{\rm sph} \sim -k_{\rm B}T \frac{L_1^{d-2} L_2^{d-2}}{R^{2(d-2)}}, \tag{19}$$

for the grounded case, and

$$F_{\rm is}^{\rm sph} \sim -k_{\rm B}T \frac{L_1^{d} L_2^{d}}{R^{2d}}, \tag{20}$$

for the isolated case, with no orientational dependence due to symmetry.

B. Two rods

The calculation of the γ tensors for a rod of length L and thickness a is more tricky. Using the cylindrical symmetry, one obtains: $\gamma_r \sim \int_0^{1/L} dk_z \int_0^{1/a} dk_\perp k_\perp^{d-2}/(k_z^2+k_\perp^2) \sim 1/L^{d-2}$ for $d \leqslant 3$, and $\sim 1/(La^{d-3})$ for $d>3$, where the z axis is parallel to the director of the cylinder, and $\perp$ denotes the remaining directions that are perpendicular to it. The second rank tensor $\gamma_{r,ij}$ is diagonal with only two independent components: $\gamma_{r,zz} \sim \int_0^{1/L} dk_z \int_0^{1/a} dk_\perp k_\perp^{d-2} k_z^2/(k_z^2+k_\perp^2) \sim 1/L^d$ for $d \leqslant 3$, and $\sim 1/(L^3 a^{d-3})$ for $d>3$, and $\gamma_{r,\perp\perp} \sim \int_0^{1/L} dk_z \int_0^{1/a} dk_\perp k_\perp^d/(k_z^2+k_\perp^2) \sim 1/(La^{d-1})$. If the unit vector $\hat{\mathbf{d}}$ denotes the director of the rod, the inverse second rank γ tensor that appears in the expression for the interaction can be written as $\gamma_{r,ij}^{-1} \sim L^d \hat{d}_i \hat{d}_j$ for $d \leqslant 3$, in the limit of small thickness. Note that in this limit, the inverse γ tensors are vanishing for $d>3$, and thus rods do not interact in these high dimensions.

Using Eqs. (17) and (18), the orientation dependent part of the interaction between two rods of lengths L_1 and L_2, and directors $\hat{\mathbf{d}}_1$ and $\hat{\mathbf{d}}_2$, which are a distance R apart, reads ($d \leqslant 3$)

$$F_{\rm gr}^{\rm rod} \sim -k_{\rm B}T \frac{L_1^{d-1} L_2^{d-1}}{R^{2(d-1)}} \left[\frac{L_1}{L_2} (\hat{\mathbf{d}}_1 \cdot \hat{\mathbf{R}}_{12})^2 + \frac{L_2}{L_1} (\hat{\mathbf{d}}_2 \cdot \hat{\mathbf{R}}_{12})^2 \right], \tag{21}$$

for the grounded case, which is pairwise additive, and

$$F_{\rm is}^{\rm rod} \sim -k_{\rm B}T \frac{L_1^{d} L_2^{d}}{R^{2d}} [\hat{\mathbf{d}}_1 \cdot \hat{\mathbf{d}}_2 - d(\hat{\mathbf{d}}_1 \cdot \hat{\mathbf{R}}_{12})(\hat{\mathbf{d}}_2 \cdot \hat{\mathbf{R}}_{12})]^2, \tag{22}$$

for the isolated case, which has a squared dipolar form and is not pairwise additive.

C. Two disks

The γ tensors for a disk of radius L and thickness a can be similarly calculated within numerical prefactors using symmetry. The zeroth rank tensor can be calculated as $\gamma_d \sim \int_0^{1/a} dk_z \int_0^{1/L} dk_\perp k_\perp^{d-2}/(k_z^2+k_\perp^2) \sim 1/L^{d-2}$, where the z-axis is normal to the disk, and $\perp$ denotes the remaining directions in the subspace of the disk. The second rank tensor $\gamma_{d,ij}$ is diagonal with only two independent components: $\gamma_{d,zz} \sim \int_0^{1/a} dk_z \int_0^{1/L} dk_\perp k_\perp^{d-2} k_z^2/(k_z^2+k_\perp^2) \sim 1/(aL^{d-1})$, and $\gamma_{d,\perp\perp} \sim \int_0^{1/a} dk_z \int_0^{1/L} dk_\perp k_\perp^d/(k_z^2+k_\perp^2) \sim 1/L^d$. If we denote the unit vector perpendicular to the disk by $\hat{\mathbf{n}}$, the inverse second rank γ tensor that appears in the expression for the interaction can be written as $\gamma_{d,ij}^{-1} \sim L^d(\delta_{ij} - \hat{n}_i \hat{n}_j)$, in the limit of small thickness.

Using Eqs.(17) and (18), the orientation dependent part of the interaction between a disk of radius L_1 and normal vector $\hat{\mathbf{n}}_1$, and another one with radius L_2 and normal vector $\hat{\mathbf{n}}_2$ that is a distance R apart, reads

$$F_{\rm gr}^{\rm disk} \sim -k_{\rm B}T \frac{L_1^{d-1} L_2^{d-1}}{R^{2(d-1)}} \times \left\{ \frac{L_1}{L_2} [1 - (\hat{\mathbf{n}}_1 \cdot \hat{\mathbf{R}}_{12})^2] + \frac{L_2}{L_1} [1 - (\hat{\mathbf{n}}_2 \cdot \hat{\mathbf{R}}_{12})^2] \right\}, \tag{23}$$

for the grounded case, which is pairwise additive, and

$$\begin{aligned} F_{\rm is}^{\rm disk} \sim -k_{\rm B}T \frac{L_1^{d} L_2^{d}}{R^{2d}} & [d^2 - d - 2 + (\hat{\mathbf{n}}_1 \cdot \hat{\mathbf{n}}_2)^2 + (2d - d^2) \\ & \times (\hat{\mathbf{n}}_1 \cdot \hat{\mathbf{R}}_{12})^2 + (2d - d^2)(\hat{\mathbf{n}}_2 \cdot \hat{\mathbf{R}}_{12})^2 - 2d(\hat{\mathbf{n}}_1 \cdot \hat{\mathbf{R}}_{12}) \\ & \times (\hat{\mathbf{n}}_2 \cdot \hat{\mathbf{R}}_{12})(\hat{\mathbf{n}}_1 \cdot \hat{\mathbf{n}}_2) + d^2 (\hat{\mathbf{n}}_1 \cdot \hat{\mathbf{R}}_{12})^2 (\hat{\mathbf{n}}_2 \cdot \hat{\mathbf{R}}_{12})^2], \end{aligned} \tag{24}$$

for the isolated case, which has a squared dipolar form and is not pairwise additive.

IV. CRITICAL FLUIDS

As mentioned above, interactions could be induced between objects that modify thermal fluctuations of an order-parameter field for a critical binary mixture or a magnetic system [14]. In this case, two kinds of boundary conditions are usually considered: (i) the *ordinary* boundary condition that suppresses the order parameter at the boundary, and thus does not break its symmetry, and (ii) the *symmetry breaking* boundary condition, which sets a nonvanishing value for the order parameter at the boundary. Note that the ordinary boundary condition is the same as the grounded boundary condition in the electrostatic terminology, and that it is dif-

ficult to imagine an analog of the isolated boundary condition in these systems [17].

The fluctuations in a critical fluid are characterized by the universality class of the system. For the case when the fluid can be described by a Gaussian Hamiltonian as given in Eq. (1), all of the above results for the grounded manifolds hold for the case of the ordinary boundary condition. The interaction between manifolds with symmetry-breaking boundary conditions, where the value of the order parameter is set to Φ_α on the αth manifold, is calculated as

$$F_{\rm sb} = -k_{\rm B}T \ln \mathcal{Z}[\phi_\alpha = \Phi_\alpha], \tag{25}$$

where $\mathcal{Z}[\phi_\alpha]$ is given by Eq. (16). One obtains [18]

$$F_{\rm sb}^{\rm Gauss} = -\frac{k_{\rm B}T}{2} \sum_{\alpha \neq \beta} \left[\frac{K \Phi_\alpha \Phi_\beta \gamma_\alpha^{-1} \gamma_\beta^{-1}}{S_d (d-2) R_{\alpha\beta}^{d-2}} \right] + O(1/R^{2d-4}). \tag{26}$$

It is important to note that this interaction is independent of the orientations of the manifolds, and that the leading-order orientation dependent term for the symmetry-breaking boundary condition is the same as the case of ordinary boundary condition, and is given as in Eq. (17). This interaction is orientationally pairwise additive.

For a nontrivial universality class, one should make use of more complicated Hamiltonians with nonlinear terms. It is then possible to calculate the Casimir energy expressions using field-theoretical techniques [4]. The interaction between two spheres in an arbitrary critical system has in fact been calculated exactly in Ref. [19] using conformal-invariance methods. The interaction for the case of symmetry-breaking boundary conditions (on both spheres) is obtained as $1/R^{d-2+\eta}$ [20], while for the case of ordinary boundary conditions it is found as $1/R^{2(d-1/\nu)}$, where η and ν are critical exponents of the system [19]. One can easily check that for the case of the Gaussian universality class, where $\eta = 0$ and $\nu = 1/2$, they coincide with the results of Eqs. (26) and (17).

It is interesting to note that the power law for the symmetry-breaking case is given by the two-point correlation function of the field (the spin-spin correlation in magnetic terminology), while the one for the ordinary case is given by the four-point correlation function (the energy-energy correlation) [19]. Guided by this, one can think of an effective Gaussian Hamiltonian of the form

$$\mathcal{H}_{\rm cf}[\phi] = \frac{K}{2} \int \frac{d^d \mathbf{q}}{(2\pi)^d} q^{2-\eta} |\phi(\mathbf{q})|^2, \tag{27}$$

which yields a correct form for the two-point function, and calculate the fluctuation-induced interactions using the above methods [7]. However, although it yields a correct result for the symmetry-breaking case (almost by construction), it gives a corresponding form for the ordinary case as $1/R^{2(d-2+\eta)}$ that is not correct. The reason is that the above effective Gaussian Hamiltonian does not give a correct four-point correlation function. However, it can be constructed to do so by using $q^{1/\nu}$ instead of $q^{2-\eta}$ in Eq. (27).

V. CONCLUSION

The analysis that is presented here is aimed at emphasizing the crucial role of the type of boundary conditions on fluctuation-induced interactions. Unlike the case of extended objects at close separations, where different types of boundary conditions all lead to the same form of interaction, we found that for objects at large separations, the type of boundary conditions determine the very form of the interactions, and, interestingly, whether or not they are pairwise additive.

We finally note that in addition to the classic case of van der Waals interaction between conductors in vacuum, the grounded and isolated boundary conditions have also found applications in the case of fluctuation-induced interactions in elastic media, where they lead to interesting effects [10,15,16].

ACKNOWLEDGMENTS

I am grateful to M. Kardar for invaluable discussions and comments. This research was supported in part by the National Science Foundation under Grant Nos. PHY94-07194 and DMR-98-05833.

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faces, while it reaches a constant value away from them. The space dependence of κ thus hampers the fluctuations of ϕ nearby the charged bodies, and hence effectively acts as a boundary condition on the fluctuating field. This leads to Casimir-type fluctuation-induced interactions in charged fluids [5].

[13] The salt concentration should be so low that the Debye screening length in the bulk is much larger than the distance between the external bodies that are immersed in the fluid. If this condition is not satisfied, one should use the Hamiltonian $\mathcal{H}=(K/2)\int d^d\mathbf{x}[(\nabla\phi)^2+\kappa^2\phi^2]$ instead. Consequently, the resulting fluctuation-induced interactions will be screened by the Debye length κ^{-1}.

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