Universality of the thermodynamic Casimir effect

Daniel Dantchev,[1,2,3] Michael Krech,[2,3] and S. Dietrich[2,3]
[1]*Institute of Mechanics, BAS, Academic Georgy G. Bonchev St. bl. 4, 1113 Sofia, Bulgaria*
[2]*Max-Planck-Institut für Metallforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany*
[3]*Institut für Theoretische und Angewandte Physik, Universität Stuttgart, Pfaffenwaldring 57, D-70569 Stuttgart, Germany*
(Received 23 September 2002; published 27 June 2003)

Recently a nonuniversal character of the leading spatial behavior of the thermodynamic Casimir force has been reported [X. S. Chen and V. Dohm, Phys. Rev. E **66**, 016102 (2002)]. We reconsider the arguments leading to this observation and show that there is no such leading nonuniversal term in the systems with short-ranged interactions if one treats properly the effects generated by a sharp momentum cutoff in the Fourier transform of the interaction potential. We also conclude that lattice and continuum models then produce results in mutual agreement independent of the cutoff scheme, contrary to the aforementioned report. All results are consistent with the *universal* character of the Casimir force in the systems with short-ranged interactions. The effects due to dispersion forces are discussed for the systems with periodic or realistic boundary conditions. In contrast to the systems with short-ranged interactions, for $L/\xi \gg 1$, one observes leading finite-size contributions governed by power laws in L due to the subleading long-ranged character of the interaction, where L is the finite system size and ξ is the correlation length.

DOI: 10.1103/PhysRevE.67.066120 PACS number(s): 64.60.Fr, 75.40.−s

I. INTRODUCTION

According to our present understanding, the Casimir effect is a phenomenon common to all systems characterized by fluctuating quantities on which external boundary conditions are imposed.

The confinement of quantum-mechanical vacuum fluctuations of the electromagnetic field causes long-ranged forces between two conducting uncharged plates, which is known as the (quantum-mechanical) Casimir effect [1–4]. The corresponding force between the plates is called the Casimir force. In this form, the phenomenon was predicted in 1948 [1] by Casimir.

The confinement of critical fluctuations of an order parameter also induces long-ranged forces between the system boundaries [5–7]. This is known as the statistical-mechanical (thermodynamic) Casimir effect. In this form, the effect was discussed by Fisher and de Gennes [5] already in 1978.

The Casimir forces arise from the influence of one portion of a system, via fluctuations, on another portion some distance away.

The best known example of a Casimir force is the van der Waals interaction between neutral molecules. In this case, the correlations between the fluctuations are mediated by photons, i.e., massless excitations of the electromagnetic field. When the system is a thermodynamic one, important examples of such massless excitations include Goldstone bosons and order parameter fluctuations at critical points.

The quantum-mechanical Casimir effect has been experimentally verified with impressive experimental precision [8] (see also Refs. [9] and [10]). One uses atomic force microscope techniques and measures the force between a metallized sphere and a plate. Since it turns out to be very difficult to keep two plates parallel with the required accuracy, there is only one recent experiment [11] in which the original theoretical parallel plate setup as studied by Casimir was used. In this experiment, it has been found that the measured Casimir force agrees with the predicted one within 15% accuracy. It is interesting to note that at distances of the order of 10 nm between the plates, the force produces a pressure of the order of 1 atm. Therefore, the Casimir effect is considered to be very important for the design of nanoscale devices (see, e.g., Ref. [12] and references therein).

In statistical mechanics, the Casimir force is usually characterized by the excess free energy coming from the *finite-size contributions* to the free energy of the system. The parallel plate or film geometry turns out to be of great practical importance for the experimental setups.

A useful model for the investigation of generic finite-size effects is given by an $O(n)$-symmetric spin system ($n \geq 1$), confined to a film geometry ($L \times \infty^2$) with periodic boundary conditions τ. Models of this sort serve as theoretical descriptions of magnets or fluids confined between two parallel plates of infinite area. The Casimir force per unit area in these systems is defined as

$$F^{\tau}_{Casimir}(T,L) = -\frac{\partial f^{\tau}_{\mathrm{ex}}(T,L)}{\partial L}, \tag{1}$$

where $f^{\tau}_{\mathrm{ex}}(T,L)$ is the excess free energy

$$f^{\tau}_{\mathrm{ex}}(T,L) = f^{\tau}(T,L) - L f_{\mathrm{bulk}}(T). \tag{2}$$

Here $f^{\tau}(T,L)$ is the full free energy per unit area (and per $k_B T$) of such a system and f_{bulk} is the bulk free energy density.

According to the definition given by Eq. (1), the thermodynamic Casimir force is a generalized force conjugate to the distance L between the boundaries of the system with the property $F^{\tau}_{\mathrm{Casimir}}(T,L) \rightarrow 0$ for $L \rightarrow \infty$. We are interested in the behavior of $F^{\tau}_{\mathrm{Casimir}}$ when $L \gg a$, where a is a typical microscopic length scale. In this limit, finite-size scaling theory is applicable. The *sign* of the Casimir force is of particular interest. It is supposed that if the boundary conditions

τ are the same at both surfaces, $F^{\tau}_{\text{Casimir}}$ will be *attractive* [13,14,16] (strictly speaking, for an Ising-like system this should hold above the wetting transition temperature T_w [13,14,17]). In the case of a fluid confined between identical walls, this implies an attractive force between the walls for large separations. When the boundary conditions *differ* between the confining surfaces, the Casimir force is expected to be *repulsive* [13,15,16]. The current experimental situation will be discussed later in the paper. Here we only mention that these experiments are in qualitative and, in some cases, even in quantitative agreement.

In this paper, we discuss the behavior of the thermodynamic Casimir force in systems with short ranged and with subleading long-ranged (dispersion) forces, which are present, e.g., in real fluids. Both interactions lead to the same universality class, provided that the dimensionality d of the system and the symmetry of the ordered state are the same. Despite this similarity we will see that, in comparison with systems with short-ranged forces, new important finite-size contributions exist in systems with dispersion forces. We shall also discuss proper boundary conditions for the systems with subleading long-ranged interactions, and we shall reconsider several recent statements [18] for the behavior of the finite-size free energy and the Casimir force in the systems with short-ranged interactions and with dispersion forces.

From the definition in Eqs. (1) and (2), it is clear that one needs to know the critical behavior of the free energy in a slab geometry in order to derive the behavior of the Casimir force. Based on numerous investigations, it has turned out that the thermodynamic behavior of a system near a second-order phase transition exhibits scale invariance and universality [19,20]. In order to set the stage for our considerations, we first recall certain bulk properties.

A. Bulk systems

Scale invariance and universality hold for the singular part of a thermodynamic function. For later reference, we quote the decomposition into a regular and a singular part of the free energy f in units of k_BT_c and per unit volume of, e.g., an Ising ferromagnet:

$$\begin{aligned} f(t,h) &= f_{reg}(t,h) + f_{sing}(t,h) \\ &= f_{reg}(t,h) + |t|^{2-\alpha} A_1 F_{\pm}(A_2 h |t|^{-\Delta}), \end{aligned} \tag{3}$$

where $t=(T-T_c)/T_c \gtrless 0$ is the reduced temperature, h is the external magnetic field, Δ is the critical exponent associated with the magnetic field, α is the critical exponent of the specific heat, A_1 and A_2 are nonuniversal (system dependent) metric factors, and $F_{\pm}$ are universal scaling functions.

In the scaling limit, the two-point correlation function in zero field, which is of particular interest in the present context, has the form [21,22]

$$G(r,t\gtrless 0) = D r^{-(d-2+\eta)} g_{\pm}(r/\xi_{\pm}) \tag{4}$$

with $\xi_{\pm}$ as the correlation length given by

$$\xi_{\pm}(t,h=0) = \xi_0^{\pm}|t|^{-\nu}, \quad t\to 0. \tag{5}$$

The nonuniversal constants D and ξ_0^+ can be related to ξ_0^-, A_1, and A_2 via $\xi_0^+/\xi_0^- = Q_1$, $A_1 = Q_2(\xi_0^+)^{-d}$, and $A_2 = Q_3\sqrt{D}(\xi_0^+)^{(d+2-\eta)/2}$ with Q_1, Q_2, and Q_3 being universal, which leads to the hyperuniversality hypothesis in the form of two-scale factor universality [23]. In the form given above, Eqs. (4) and (5) are valid for Ising-like systems only. For $O(n)$ models, $n\geqslant 2$, one has in addition to take into account that $\xi_-(t\leqslant 0)\equiv\infty$. The universal scaling function $g_+(x)$ decays exponentially for $x \gtrsim 1$. The physical origin for the onset of scale invariance can be traced back to the divergence of the correlation length $\xi_{\pm}$. Consensus has emerged that these statements hold in systems governed by short-range interaction potentials, i.e., decaying exponentially or being of a finite range.

If the exchange interaction in an Ising model on a lattice in d dimensions decays algebraically,

$$J(\mathbf{r}) = \frac{J}{1+(r/a)^{d+\sigma}}, \quad r\equiv|\mathbf{r}|\geqslant a>0, \tag{6}$$

where a is the lattice constant, the value of the decay exponent σ is crucial with respect to universality. For $\sigma>2$, the leading thermodynamic critical behavior is characterized by the critical exponents and scaling functions for short-ranged interactions [24]. Mean-field theory holds for $d>d_c=4$ irrespective of the value of σ. For $\sigma<2$, the upper critical dimension is reduced to $d_c(\sigma)=2\sigma$ [24,25], and the values of the critical exponents depend on σ for $\sigma<d<d_c(\sigma)$ [24,26,27]. The crossover from short-ranged to long-ranged critical behavior occurs for $\sigma=2-\eta_{sr}(d)$, where $\eta_{sr}(d)$ is the critical exponent for the short-ranged system (for a given fixed spatial dimension d) [28–32]. This crossover has recently been reexamined numerically in $d=2$ in Ref. [33].

Fluids are governed by dispersion forces. In the sense of Eq. (6), dispersion (van der Waals) forces in $d=3$ dimensions are characterized by $\sigma=3$ in the nonretarded case and by $\sigma=4$ in the retarded case. Therefore, the leading thermodynamic critical behavior of a fluid is characterized by critical exponents and scaling functions for short-ranged interactions and the contributions due to the power-law decay of the interaction potential lead to corrections to the asymptotic scaling behavior. Thus for $\sigma>2$ we refer to interaction potentials governed by Eq. (6) as subleading long-ranged interactions.

As an illustration of this case, Fig. 1 displays schematically the two-point correlation function $G(r,t)$. The universal decay of $G(r,t)$ is governed by Eq. (4) only within the critical regime. For distances r smaller than the lower limit of this regime, generically nonuniversal microscopic effects govern the behavior of the correlation function. For distances larger than the upper limit of this regime, the interaction potential itself governs the further decay of the correlation function. Note that $r^*/\xi\to\infty$ as $T\to T_c$, i.e., the critical regime expands as the critical point is approached [35–37].

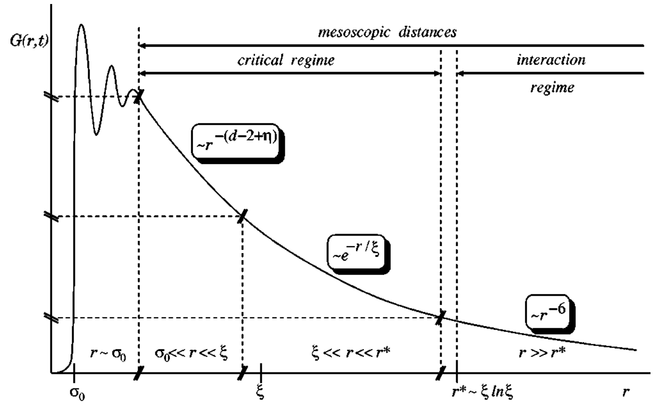

FIG. 1. Schematic view of the density-density correlation function $G(r,t)$ in a fluid governed by dispersion forces in $d=3$. The behavior is shown on various length scales, where the tilted double slashes // indicate breaks in scale. For microscopic distances of the order of the particle diameter σ_0, "packing" effects lead to oscillations decaying exponentially [34]. Beyond a crossover regime (not shown), the correlation function decays according to the power law given in Eq. (4) as long as $r \ll \xi$. Beyond another crossover (not shown) the further decay is exponential for $r \gg \xi$ which finally crosses over to the interaction dominated regime for $r > r^*$, where the ultimate decay of the correlation function follows the decay of the interaction potential $\sim r^{-6}$ in the nonretarded regime and $\sim r^{-7}$ in the retarded regime (not shown). The behavior $r^* \sim \xi \ln \xi$ of the crossover distance r^* [35–37] illustrates that r^* diverges more strongly than the correlation length ξ in the vicinity of T_c, i.e., the critical regime, in which the universal properties hold, expands as T approaches T_c.

The widening of the critical regime leads to the divergence of the compressibility $\kappa(t) \sim \int_0^\infty dr\, r^2 G(r,t) \sim |t|^{-\gamma}$, for $t \to 0$.

For short-ranged interactions, one has to bear in mind that Eq. (4) is also only valid within a critical regime $r < r^*_{\rm sr}$. Exact results for the two-dimensional (2D) Ising model [38] and mean-field results [39] suggest that $r^*_{\rm sr} \sim \xi^2$, whereas the spherical model yields the estimate $r^*_{\rm sr} \sim \xi^3$ [40]. For $r \gg r^*_{\rm sr}$, the correlation function decays again exponentially, but it contains a nonuniversal prefactor [40], i.e., the *leading-order* behavior becomes nonuniversal. This demonstrates that in the case of short-ranged interactions, the width of the critical regime is much larger than for a corresponding system with subleading long-ranged interactions.

B. Finite-size scaling

Finite-size scaling asserts that near the bulk critical temperature T_c, the influence of a finite sample size L on the critical phenomena is governed by universal finite-size scaling functions that depend on the ratio L/ξ, so that the rounding of the thermodynamic singularities sets in for $L/\xi \simeq O(1)$ [7,41–45].

From the above discussion of the behavior of $G(r,t)$, one expects that the deviations from standard finite-size scaling behavior will be observed for $L \gg r^*$, where r^* is a crossover length with the property $r^* \gg \xi$. In particular, one has $r^* \sim \xi^2$ for the Ising model or within mean-field theory, $r^* \sim \xi^3$ for the spherical model [40], and $r^* \sim \xi \ln \xi$ for subleading long-ranged interactions [37,46].

This view has recently been challenged by Chen and Dohm [18], who purportedly report, for the systems with short-ranged interactions, leading finite-size contributions different from the ones expected from the above discussion. This would invalidate the current understanding of finite-size scaling. In particular, these results can lead to the expectation of a *nonuniversal* Casimir force at T_c for a fluid between parallel plates at a distance L. If correct this would be of major theoretical [6,48,49] and experimental interest [50–52]. Specifically, based on the exact results for the $O(n)$-symmetric ϕ^4 field theory in the large-n limit (mean spherical model), the authors of Ref. [18] report the following result for the singular part $f_s(t,L,\Lambda)$ of the finite-size contribution $f(t,L,\Lambda)$ to the free energy density of a system with periodic boundary conditions and purported short-ranged interactions in $2<d<4$:

$$f_s(t,L,\Lambda) = L^{-2}\Lambda^{d-2}\Phi(\xi^{-1}\Lambda^{-1}) + L^{-d}X^{sr}(L/\xi). \quad (7)$$

The parameter Λ is an ultraviolet momentum cutoff and the function Φ has the property $\Phi(0)>0$. For the Casimir force defined by

$$F_{Casimir}(t,L,\Lambda) \equiv -\frac{\partial}{\partial L}\{L[f(t,L,\Lambda) - f(t,\infty,\Lambda)]\}, \quad (8)$$

Eq. (7) implies a leading nonuniversal (cutoff dependent) nonscaling term $\sim L^{-2}$ in the behavior of the Casimir force because the scaling function $X^{sr}(x) \sim \exp(-x)$ when $x \gg 1$ [7,41–43,45]. Therefore Eqs. (7) and (8) would also imply *nonuniversal* Casimir amplitudes

$$\Delta_{Casimir}(d) = \Lambda^{d-2}\Phi(0) \quad (9)$$

in $d>2$ dimensions.

In the following, we shall show that the results reported in Ref. [18] can be traced back to using a peculiar model in which the interactions are neither short ranged nor of the subleading long ranged type, so that the model does not relate to any physical realization. We find that if the periodicity and analyticity of the Fourier transform $J(\mathbf{k})$ of the interaction $J(r)$ at the boundary of the Brillouin zone (in the case of a lattice model), and the analyticity of $J(\mathbf{k})$ at the cutoff $k=\Lambda$ (in the case of an off-lattice model) are preserved in the theoretical analysis, then the L^{-2} term in Eq. (7) *vanishes identically*. We also show that the presence or absence of this term does *not* depend on the range of the interactions. If the above requirements for $J(\mathbf{k})$ at the boundary of the Brillouin zone or at $k=\Lambda$ are violated, a corresponding nonuniversal, nonscaling term of order L^{-2} will be observed in the finite-size behavior of *any* thermodynamic function. A discussion on the influence of the cutoff on the finite-size behavior of the susceptibility has already been presented in Ref. [46]; see also the "note added in proof" of Ref. [54]. In Secs. II and III, we present a general and unified approach that is designed to avoid similar artificial effects; this should be useful also in the context of quantum phase transitions and field

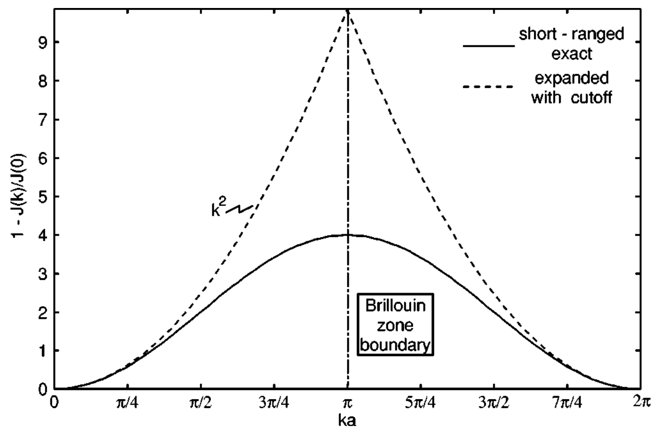

FIG. 2. Dispersion relation $1-J(k)/J(0)$ as function of k for nearest-neighbor interactions in one dimension (solid line) in comparison with a k^2 spectrum with a sharp cutoff at the Brillouin zone boundary, for $d=1$ dimension, as implemented in Ref. [18] (dashed line). The zone boundary is marked by the vertical dash-dotted line and a is the lattice constant. Note that the application of a sharp cutoff to a pure k^2 spectrum in the first Brillouin zone implies an artificial cusplike nonanalyticity of the dispersion relation at the zone boundaries. This is absent for the genuine short-ranged interactions.

theory. In Sec. III, we also summarize the present state of knowledge on the finite-size behavior of the systems with subleading long-range interactions, focusing on the expected behavior of the singular part of the free energy. One should distinguish between the cases (i) $d+\sigma<6$ and (ii) $d+\sigma=6$, which contains the physically most important case of dispersion forces in $d=3$ with $\sigma=3$. In case (ii), one expects additional logarithmic finite-size contributions. The paper closes in Sec. IV with a summary and concluding remarks, where we also discuss possible boundary conditions for systems with subleading long-ranged interactions.

II. FINITE-SIZE BEHAVIOR OF THE FREE ENERGY DENSITY

In this section, we present our critique of the finite-size scaling analysis of the free energy and the Casimir force presented in Ref. [18]. As pointed out already, the statements of Ref. [18] are based on the exact results for the $O(n)$-symmetric ϕ^4 field theory in the large-n limit (mean spherical model) with periodic boundary conditions.

A. Analytical properties

Before we turn to the finite-size analysis, we discuss briefly the consequences of the assumptions for the Fourier transform of the interaction $J(\mathbf{k})$ used in Ref. [18] for the *bulk* properties of the model. For a system on a lattice, $J(\mathbf{k})=\Sigma_{\mathbf{r}}J(\mathbf{r})\exp(i\mathbf{k}\cdot\mathbf{r})$, where the sum runs over the lattice sites. For an off-lattice system, the sum has to be replaced by the corresponding integral.

In Fig. 2, we compare $J(\mathbf{k})$ for a short-ranged (nearest-neighbor) lattice model [see, cf. Eq. (22)] with the standard k^2 spectrum in the infrared limit for short-ranged interactions in the spirit of Ref. [18] endowed with a cutoff $\Lambda=\pi/a$ at the boundaries $\pm\pi/a$ of the first Brillouin zone in one dimension. The restriction to one dimension only simplifies the notation, and is not essential for the following arguments. From Fig. 2, it is obvious that the continuation of the k^2 spectrum, which is correct only in the infrared limit $k\to 0$, to the full Brillouin zone and its truncation at the zone boundaries introduces a cusplike nonanalyticity into the spectrum. This nonanalyticity is artificial and is not a generic feature of the short-ranged interactions. As we shall demonstrate below, this artificial choice is the reason for the nonscaling and nonuniversal finite-size effects $\sim L^{-2}$ reported in Ref. [18]. In fact, the importance of the properties of the dispersion relation at the Brillouin zone boundary for the critical finite-size behavior of the free energy in $d=2$ dimensions has already been mentioned by Cardy [see Eq. (3.12) of Ref. [55]] in the course of deriving the Casimir amplitude $\Delta_{Casimir}(2)=-\pi c/6$ in $d=2$ the for periodic boundary conditions, where c is the central charge of the model under consideration (e.g., $c=1/2$ for the critical 2D Ising model with short-ranged interactions).

It is instructive to investigate the consequences of a nonanalytic spectrum of the kind displayed in Fig. 2 in real space. According to Ref. [18], the corresponding correlation function for $r\gg\xi$ in d dimensions reads

$$G(r,t)=2\Lambda^{d-2}(2\pi r\Lambda)^{-(d+1)/2}\frac{\sin[\Lambda r-\pi(d-1)/4]}{1+\xi^{-2}\Lambda^{-2}} + O(\exp(-r/\xi)). \tag{10}$$

Thus the correlation function decays according to a power law with the decay exponent $(d+1)/2$ rather than exponentially. Furthermore, the correlation function oscillates with a period set by the inverse of the cutoff Λ. For separations $r>r^*$ (see Fig. 1), Eq. (10) therefore implies that the interaction potential for a system with a truncated k^2 spectrum, as shown in Fig. 2, should not only be *leading long ranged* rather than *short ranged* or even *subleading long ranged* in $2\leqslant d\leqslant 4$, but also containing both positive and negative contributions. Therefore, for all spatial dimensions of physical relevance, the model investigated in Ref. [18] would appear to correspond to a model with competing leading long-ranged interactions in real space. For such a model, the finite-size scaling as developed for the systems with short-ranged or subleading long-ranged interactions is not expected to be applicable.

B. Finite-size properties of the $O(n\to\infty)$ model

We substantiate our view by turning to a detailed analysis of the finite-size behavior of the free energy and other thermodynamic functions, i.e., we provide an account of the mathematical mechanism that produces the nonuniversal and non-scaling leading finite-size effects reported in Ref. [18].

As a case study, we quote the expression for the total free energy density of a fully finite mean spherical model [the $n\to\infty$ limit of an $O(n)$ model] with nearest-neighbor interactions of strength J on a hypercubic lattice, which is given by [7]

$$\beta f(K,h|\mathbf{L},\mathbf{\Lambda})=\frac{1}{2}\sup_{\tau>0}\left\{-\frac{h^2}{K\tau}+\frac{1}{L_1\times\cdots\times L_d}\times\sum_{\mathbf{k}}\ln\left[\tau+\sum_{j=1}^{d}2(1-\cos k_j)\right]-K\tau\right\}+\frac{1}{2}\left[\ln\frac{K}{2\pi}-2dK\right], \quad (11)$$

where $K=\beta J$ and h is a properly normalized magnetic field. The parameter τ is related to the correlation length ξ via $\tau=\xi^{-2}$ [47] which relates τ also to the reduced temperature t. The relevant physical information of Eq. (11) is contained in the sum over the wave vectors $\mathbf{k}=(k_1,\ldots,k_d)$ in the first Brillouin zone of the simple cubic lattice. For general interaction potentials on general lattices, this sum involves the dispersion relation $\omega(\mathbf{k})$, where, e.g., $\omega(\mathbf{k})=\Sigma_{j=1}^{d}2(1-\cos k_j)$ for the nearest-neighbor interactions on a simple cubic lattice [see Eq. (11) and, cf. Eq. (22)]. In order to provide a general description of finite-size scaling of the free energy, we will therefore consider the quantity [7] [see also Eq. (14) in Ref. [18]]

$$U_{d,\sigma}(\tau,\mathbf{L},\mathbf{\Lambda})=\frac{1}{2L_1\times\cdots\times L_d}\sum_{\mathbf{k}\in\mathcal{B}}\ln[\tau+\omega(\mathbf{k})] \quad (12)$$

as a function of the system size $\mathbf{L}$, where $\mathbf{L}=(L_1,\ldots,L_d)$ denotes the set of lengths that determine the geometry of the system, $\mathbf{\Lambda}=(\Lambda_1,\ldots,\Lambda_d)$ is the set of cutoffs in reciprocal (i.e., k) space, and τ is a quantity proportional to the reduced temperature t, e.g., $\tau=\xi^{-2}$ for the mean spherical model with short-range interactions. For lattice systems, $\mathbf{k}$ belongs to the first Brillouin zone $\mathcal{B}$ and if the system is on a hypercubic lattice, one has $\Lambda_i=\pi/a_i$, where a_i is the lattice spacing along the direction i, $i=1,\ldots,d$. For off-lattice systems the summation is carried out over those values, $\mathbf{k}\in\mathcal{B}$, that fulfill the requirements $-\Lambda_i\leqslant k_i<\Lambda_i$, $i=1,\cdots,d$. However, for off-lattice systems, there is no obvious choice for the cutoff. One usually takes $\Lambda\simeq\tilde{a}^{-1}$, where $\tilde{a}$ is some fixed characteristic microscopic length of the system. For a fluid, $\tilde{a}$ can be taken to be the diameter σ_0 of a fluid particle (see also Fig. 1). In Eq. (12), the subscript σ characterizes the range of the interaction. The fluctuation spectrum $\omega(\mathbf{k})$ of the order parameter is given as a linear function of the Fourier transform $J(\mathbf{k})$ of the interaction [see, cf. Eqs. (22) and (23)].

The formal expression given by Eq. (12), on which our specific considerations in this section are based, has widespread applications. The expression in Eq. (12) always appears as the one-loop contribution to the free energy in field-theoretic Ginzburg-Landau models [48]. The line of arguments presented here is therefore of general importance, and is not limited to the specific model under consideration here.

For periodic boundary conditions, $L_i\Lambda_i/(2\pi)\equiv M_i$ are integer numbers, $i=1,\ldots,d$, where $\Pi_{i=1,\ldots,d}M_i\equiv N$ fixes the number of degrees of freedom in the system. The values of the components k_i of the vector $\mathbf{k}$ are given by $k_i=2\pi m_i/L_i$, with $-M_i\leqslant m_i\leqslant M_i-1$, $i=1,\cdots,d$.

In order to analyze the sum in Eq. (12), we use the Poisson summation formula

$$\sum_{m=a}^{b}f(m)=\sum_{n=-\infty}^{\infty}\int_a^b dm e^{i2\pi mn}f(m)+\frac{1}{2}[f(a)+f(b)]. \quad (13)$$

After some algebra, one obtains

$$U_{d,\sigma}(\tau,\mathbf{L},\mathbf{\Lambda})=U_{d,\sigma}(\tau,\mathbf{\Lambda})+\Delta U_{d,\sigma}(\tau,\mathbf{L},\mathbf{\Lambda}), \quad (14)$$

where

$$U_{d,\sigma}(\tau,\mathbf{\Lambda})=\frac{1}{2(2\pi)^d}\int_{-\Lambda_1}^{\Lambda_1}dm_1\cdots\int_{-\Lambda_d}^{\Lambda_d}dm_d\ln[\tau+\omega(\mathbf{m})] \quad (15)$$

takes into account the contributions of the bulk system, while

$$\Delta U_{d,\sigma}(\tau,\mathbf{L},\mathbf{\Lambda})=\frac{1}{2(2\pi)^d}\sum_{\mathbf{n}\neq\mathbf{0}}\int_{-\Lambda_1}^{\Lambda_1}dm_1\cdots\int_{-\Lambda_d}^{\Lambda_d}dm_d\times\exp\left(i\sum_{j=1}^{d}n_jm_jL_j\right)\ln[\tau+\omega(\mathbf{m})] \quad (16)$$

incorporates all contributions due to the finite size of the system. For further analysis of the Casimir effect, the film geometry $L\times\infty^{d-1}$ is the most relevant one. It is obtained from Eq. (16) in the limit $L_2\to\infty,\ldots,L_d\to\infty$, setting $L_1\equiv L$. In order to simplify the notation, we finally set $\Lambda_1=\Lambda_2=\cdots=\Lambda_d\equiv\Lambda$ so that

$$\Delta U_{d,\sigma}(\tau,L,\Lambda)=\frac{1}{2(2\pi)^d}\sum_{n_1\neq 0}\int_{-\Lambda}^{\Lambda}dm_1\cdots\int_{-\Lambda}^{\Lambda}dm_d\times\exp(in_1m_1L)\ln[\tau+\omega(\mathbf{m})], \quad (17)$$

which after two integrations by parts with respect to m_1 can be rewritten as

$$\Delta U_{d,\sigma}(\tau,L,\Lambda)=\Delta U_{d,\sigma}^{(1)}(\tau,L,\Lambda)-L^{-2}\frac{1}{2(2\pi)^d}\times\sum_{n_1\neq 0}\frac{1}{n_1^2}\int_{-\Lambda}^{\Lambda}dm_1\cdots\int_{-\Lambda}^{\Lambda}dm_d\times\exp(in_1m_1L)\partial_{m_1}\times\left[\frac{\partial_{m_1}\omega(\mathbf{m})}{\tau+\omega(\Lambda,m_2,\ldots,m_d)}\right], \quad (18)$$

with

$$\Delta U_{d,\sigma}^{(1)}(\tau,L,\Lambda)=L^{-2}\frac{\pi^2}{6}\frac{1}{(2\pi)^d}\int_{-\Lambda}^{\Lambda}dm_2\cdots\int_{-\Lambda}^{\Lambda}dm_d \times\frac{\partial_{m_1}\omega(\mathbf{m})|_{m_1=\Lambda}-\partial_{m_1}\omega(\mathbf{m})|_{m_1=-\Lambda}}{\tau+\omega(\Lambda,m_2,\ldots,m_d)}. \tag{19}$$

The above expression is obtained by the identity transformations of the initial sum, and is valid both for lattice and off-lattice systems. In the analysis below we show that it is the term $\Delta U_{d,\sigma}^{(1)}(\tau,L,\Lambda)$ which produces the contributions, on which the statements of Ref. [18] are based.

First, we evaluate this term for lattice systems. If the interactions $J(\mathbf{r})\geqslant 0$ are such that they depend only on the distances between the particles, and these are the only interactions we are concerned with here, then $J(\mathbf{k})=J(-\mathbf{k})$. Since there is no physical reason for singularities anywhere except at $k=0$, the derivatives of $J(\mathbf{k})$ with respect to $\mathbf{k}$ should exist at least for all $k\neq 0$ and therefore $\partial_{\mathbf{k}}J(\mathbf{k})=-\partial_{\mathbf{k}}J(-\mathbf{k})$. This holds for lattice and off-lattice systems. For lattice systems, $J(\mathbf{k})$ is a periodic function with the property

$$J(\mathbf{k}+2\Lambda_i\mathbf{e}_i)=J(\mathbf{k}), \tag{20}$$

where $\mathbf{e}_i$ is a unit vector in reciprocal space. This implies that $\partial_{\mathbf{k}}J(\mathbf{k})=0$ at the borders of the Brillouin zone and therefore

$$\Delta U_{d,\sigma}^{(1)}(\tau,L,\Lambda)\equiv 0. \tag{21}$$

Note that the above result does not depend on the range of the interaction—it is true for short-ranged, subleading long-ranged, as well as for leading long-ranged interactions. As an illustration of the above general arguments, we recall that the exact Fourier transform of the nearest-neighbor interaction on a d-dimensional hypercubic lattice reads

$$J(\mathbf{k})=2J\sum_{j=1}^{d}\cos k_j\equiv J[2d-\omega(\mathbf{k})], \tag{22}$$

where

$$\omega(\mathbf{k})\equiv\sum_{j=1}^{d}2(1-\cos k_j). \tag{23}$$

The general properties of $J(\mathbf{k})$ that we have discussed above can be easily verified from Eqs. (22) and (23).

We now reconsider the quantity $\Delta U_{d,\sigma}^{(1)}(\tau,L,\Lambda)$ if the exact spectrum is replaced by its asymptotic form, valid in the infrared limit $k\rightarrow 0$, for all $\mathbf{k}\in\mathcal{B}$. This is a very common procedure in the theory of critical phenomena, based on the general idea that only long wavelength (small k) contributions are important for the critical properties of the system. This leads to $\omega(\mathbf{m})=\mathbf{m}^2$, which is the spectrum used in Ref. [18]. One immediately obtains

$$\begin{aligned}\Delta U_{d,\sigma}^{(1)}(\tau,L,\Lambda)&=\frac{\Lambda}{L^2}\frac{2\pi^2}{3}\frac{1}{(2\pi)^d}\int_{-\Lambda}^{\Lambda}dm_2\cdots\int_{-\Lambda}^{\Lambda}dm_d \times\frac{1}{\tau+\Lambda^2+m_2^2+\cdots+m_d^2}\\&=\frac{\Lambda^{d-2}}{L^2}\frac{1}{6}\frac{1}{(2\pi)^{d-2}}\int_{-1}^{1}dm_2\cdots\int_{-1}^{1}dm_d \times\frac{1}{1+\tau/\Lambda^2+m_2^2+\cdots+m_d^2}.\end{aligned} \tag{24}$$

We recall that in the spherical limit of the $O(n)$ model, one has $\tau=\xi^{-2}$. Equation (24) *exactly* reproduces the nonuniversal leading but nonscaling finite-size contribution to the free energy as reported in Eq. (16) of Ref. [18] for the corresponding field-theoretic model. Similar contributions exist also for subleading long-ranged interactions to which we turn in Sec. III. According to the above considerations, such nonuniversal (cutoff dependent) contributions of the order of L^{-2} will always appear if

$$\left.\frac{\partial J(\mathbf{k})}{\partial k_1}\right|_{k_1=\Lambda}\neq\left.\frac{\partial J(\mathbf{k})}{\partial k_1}\right|_{k_1=-\Lambda}. \tag{25}$$

Note that only the properties of the Fourier transform $J(\mathbf{k})$ of the interaction at the boundary of the set $\mathcal{B}$ of allowed k values are important here. For a field-theoretic model, these are definitely a matter of definition. For lattice models, these properties follow automatically. Approximating the spectrum $\omega(\mathbf{k})$ by its infrared asymptotic behavior leads to an artificial cusplike singularity at the border of the Brillouin zone as is illustrated in Fig. 2. A corresponding approximation for Eq. (3.11) in Ref. [55] would lead to an incorrect prediction of the critical finite-size contribution to the free energy [see Eq. (3.12) in Ref. [55]] leading to a vanishing Casimir amplitude.

Before we consider how to modify the definition of the continuous field-theoretic model as to avoid a nonzero $\Delta U_{d,\sigma}^{(1)}(\tau,L,\Lambda)$, we make some general remarks. First, the considerations presented above can easily be extended to any geometry of the type $L^{d-d'}\times\infty^{d'}$, $0\leqslant d'\leqslant d$. Second, in the above discussion we did *not* specify the type of the interactions—short ranged, leading long ranged, or subleading long ranged. This implies that the L^{-2} corrections in question exist for *any* type of interaction for periodic boundary conditions, provided Eq. (25) is valid. Furthermore, further integrations by parts yield additional contributions of the order L^{-4}, L^{-6}, etc. In $2<d<4$ dimensions, only the term

L^{-2} is important, but in $d>4$ spurious finite-size terms of the orders L^{-2} and L^{-4} will be generated. Finally, we note that the precise form of the integrand in Eq. (17) was not used in the above analysis. This implies that spurious L^{-2} corrections also occur in other quantities such as the susceptibility [53,54] (see also the discussion in Appendix G of Ref. [46]), the specific heat 56, etc. The influence of different cutoff types (sharp or smooth) and of the truncation of the expansion of the Fourier transform of the interaction on the finite-size behavior of the susceptibility has been considered in detail in Ref. [46]. The only difference with respect to the free energy is that the susceptibility diverges as $L^{\gamma/\nu}$ at T_c, leaving any L^{-2} contribution as a *small* correction, whereas the singular part of the free energy behaves as L^{-d}, and therefore the cutoff dependent term of the order L^{-2} becomes *dominant* in the critical region for $d>2$.

Since there is no physical reason for the introduction of a *sharp* cutoff in the *k*-space representation of a field-theoretic model, one option to avoid artificial L^{-2} contributions is to implement of a *smooth* cutoff. Various forms of smooth cutoffs are possible [46,54]. For example, in Ref. [54] a modified continuum Ginzburg-Landau Hamiltonian has been considered (see also Ref. [57]):

$$H=\int_V d^dx\left[\frac{1}{2}r_0\varphi^2+\frac{1}{2}(\nabla\varphi)^2+u_0(\varphi^2)^2+\frac{1}{2\Lambda^2}(\nabla^2\varphi)^2\right]. \tag{26}$$

The last term in Eq. (26) introduces the smooth cutoff which is parametrized by a wave number Λ. The finite-size effects of the thermodynamic quantities differ substantially for the above Hamiltonian and the standard one with a sharp cutoff. In the framework corresponding to Eq. (26), the thermodynamic quantities approach their bulk value *exponentially* as a function of L for $T\neq T_c$ fixed, whereas for the standard Ginzburg-Landau Hamiltonian with a sharp cutoff the bulk limit is reached according to the power law $\sim L^{-2}$ for any temperature. In particular, this has been observed for the susceptibility [58], the specific heat [56], and the free energy [18]. The effect of a smooth cutoff prodecure on the finite-size behavior of the susceptibility was also discussed in detail in Ref. [46], where the aforementioned exponential finite-size behavior was recovered. As already noticed in Refs. [40,18,54], the presence of a sharp cutoff is mandatory for the occurrence of the aforementioned nonuniversal L^{-2} contributions to finite-size scaling. For general $d>2$, it was also realized that a close relationship exists between a non-exponential large-distance behavior of the bulk correlation function (generated by the sharp cutoff) and the power-law finite-size behavior of both the susceptibility above T_c [40] and the singular part of the free energy [18]. Here we have demonstrated that all such finite-size effects arise from a single origin and are unphysical mathematical artifacts due to the imposed singularity of $J(k)$ at the boundary of the allowed k values. This, in turn, generates long-ranged correlations in real space. In order to eliminate spurious finite-size contributions, we propose the replacement [see Eq. (17)]

$$\begin{aligned}\Delta U_{d,\sigma}(\tau,L,\Lambda)&\to\Delta U_{d,\sigma}(\tau,L,\Lambda)-\Delta U^{(1)}_{d,\sigma}(\tau,L,\Lambda)\\&=\frac{1}{2(2\pi)^d}\sum_{n_1\neq 0}\int_{-\Lambda}^{\Lambda}dm_1\cdots\int_{-\Lambda}^{\Lambda}dm_d\\&\quad\times\exp(in_1m_1L)\ln[\tau+\omega(\mathbf{m})]\\&\quad-L^{-2}\frac{\pi^2}{6}\frac{1}{(2\pi)^d}\int_{-\Lambda}^{\Lambda}dm_2\cdots\int_{-\Lambda}^{\Lambda}dm_d\\&\quad\times\frac{\partial_{m_1}\omega(\mathbf{m})|_{m_1=\Lambda}-\partial_{m_1}\omega(\mathbf{m})|_{m_1=-\Lambda}}{\tau+\omega(\Lambda,m_2,\ldots,m_d)}\end{aligned} \tag{27}$$

and the corresponding replacements generated by the derivatives of Eq. (27) with respect to the parameter τ in the definition of each model system *regardless* of the implementation of a sharp cutoff. Within such a scheme, the well-established methods for field-theoretic calculations in the presence of sharp cutoff are preserved. For lattice system, this is an identity transformation because $\Delta U^{(1)}_{d,\sigma}(\tau,L,\Lambda)\equiv 0$ as expounded above. Note that the replacements given by Eq. (27) and its derivatives with respect to τ do not interfere with the treatment of bulk systems, since $\Delta U^{(1)}_{d,\sigma}(\tau,L=\infty,\Lambda)=0$. They only become important for studies of the finite-size scaling behavior of systems endowed with a sharp cutoff. These replacements remove the artificial cutoff dependent finite-size contributions to the thermodynamic quantities in the spherical limit $n\to\infty$ of $O(n)$ models and to one-loop order for $O(n)$ models with finite n.

III. SYSTEMS WITH SUBLEADING LONG-RANGED INTERACTIONS

First, we briefly recall the finite-size behavior of systems with subleading long-ranged interactions.

In Refs. [37,46,59], it was shown that the susceptibility of a finite system with dispersion interactions, which decay as $r^{-d-\sigma}$ for large distances, can be written for $2<d<4$, $2<\sigma<4$, and $d+\sigma<6$ in the form

$$\begin{aligned}\chi(t,L)&=L^{\gamma/\nu}X_\chi(L/\xi,bL^{2-\sigma-\eta})\simeq L^{\gamma/\nu}[X^{sr}_\chi(L/\xi)\\&\quad+bL^{2-\sigma-\eta}X^{lr}_\chi(L/\xi)],\end{aligned} \tag{28}$$

where

$$X^{sr}_\chi(x\to+\infty)\simeq X^{sr,+}_\chi x^{-\gamma/\nu}+O(\exp(-\text{const}\,x)). \tag{29}$$

For the long-ranged part one has

$$X^{lr}_\chi(x\to+\infty)\simeq X^{lr}_{\chi,1}x^{-2\gamma/\nu+\sigma}+X^{lr}_{\chi,2}x^{-2\gamma/\nu-d}. \tag{30}$$

The amplitude b is a nonuniversal parameter that can be determined from the Fourier transform of the interaction. The first term of the asymptotic behavior of $X^{lr}_\chi(x)$ yields the bulk corrections to scaling as predicted by Kayser and Raveché [35], while the second term yields the leading finite-size correction to the susceptibility for $L/\xi \gg 1$. This second term leads to $\chi(t,L)-\chi(t,\infty) \sim t^{-d\nu-2\gamma}L^{-(d+\sigma)}$, $L/\xi \gg 1$, i.e., the finite-size corrections to the bulk behavior are governed by a *power law* rather than an exponential function of the system size L. Finally, we note that for the physically most important case $d+\sigma=6$ (e.g., $d=\sigma=3$ for nonretarded van der Waals forces in $d=3$), one finds additional logarithmic corrections in Eq. (28) [37,46], which can be incorporated by the replacement $X_\chi^{lr}(x) \rightarrow X_\chi^{lr,1}(x)\ln L + X_\chi^{lr,2}(x)$ [37].

The behavior of the susceptibility outlined above is consistent with the behavior of the bulk pair correlation function in systems with dispersion forces [37],

$$G(r,t) = r^{-(d-2+\eta)}[g_\pm^{sr}(r/\xi) + r^{-(\sigma-2+\eta)}g_\pm^{lr}(r/\xi)]. \tag{31}$$

The modified Fisher-Privman [60] finite-size scaling hypothesis for the free energy density in such systems can be cast into the form

$$\begin{aligned} f_s(t,L) &= L^{-d}X(L/\xi, bL^{2-\sigma-\eta}) \\ &\simeq L^{-d}[X^{sr}(L/\xi) + bL^{2-\sigma-\eta}X^{lr}(L/\xi)], \end{aligned} \tag{32}$$

where $X^{sr}(x \rightarrow +\infty) \simeq X^{sr,+}x^d + O(\exp(-\text{const}\, x))$ is the short-ranged contribution. For the long-ranged contribution, one expects $X^{lr}(x) \simeq X_1^{lr}x^{d+\sigma+\eta-2} + X_2^{lr}x^{\eta-2}$. The first term in the asymptotic behavior of X^{lr} yields a *new bulk correction to scaling* that is due to the subleading part of the interaction (analogous to the corresponding terms predicted by Kayser and Raveché for the susceptibility [35] and observed in spherical model calculations [46]). Its temperature dependence for $T > T_c$ is given by $t^{d\nu+(\sigma+\eta-2)\nu}$. For $L/\xi \gg 1$, the second term leads to a finite-size contribution of the form $L^{-(d+\sigma)}$. As for the finite-size scaling behavior of the susceptibility, additional logarithmic corrections have to be added to X^{lr} for $d+\sigma=6$. Finally, we note that there may also be a third *constant* contribution to the asymptotic behavior of $X^{lr}(x)$ for $x \gg 1$, which does not appear in the susceptibility and which leads to a $L^{-4}\ln L$ finite-size contribution to the free energy for $d=\sigma=3$.

Equation (32) has been derived in Ref. [18] for the case $\eta=0$, where the main focus is set on the discussion of finite-size contributions. Furthermore, the results reported in Ref. [18] apparently apply only for $d+\sigma<6$ because no logarithmic corrections were found. Thus a complete verification of Eq. (32) is still missing. We now turn to the investigation of some of the consequences of the assumptions used in the model in Ref. [18].

We suppose that the interaction potential $J(\mathbf{r})$ is of the dispersion type as defined by Eq. (6). The Fourier transform of such an interaction is

$$\begin{aligned} J(\mathbf{k}) &\simeq J(\mathbf{0})[1 - v_2k^2 + v_\sigma k^\sigma - v_4k^4 + O(k^6)] \\ &\equiv J(\mathbf{0}) - K\omega(\mathbf{k})/\beta, \end{aligned} \tag{33}$$

where $k=|\mathbf{k}|$, $4>\sigma>2$; and $J(\mathbf{0})$, v_2, v_σ, and v_4 are nonuniversal positive constants. The constant $J(\mathbf{0})$ is the ground state energy of the system and $\omega(\mathbf{k}) \simeq k^2 - bk^\sigma + ck^4 + O(k^6)$, where $K=\beta v_2 J(\mathbf{0})$, $b=v_\sigma/v_2>0$, and $c=v_4/v_2>0$ are nonuniversal constants. Since $J(\mathbf{r}) \geq 0$ and thus $J(\mathbf{0})>J(\mathbf{k})$ for $\mathbf{k}\neq\mathbf{0}$, the values of b and c are such that there are no real roots of the equation $1-bk^{\sigma-2}+ck^2=0$ with respect to k.

The free energy of an $O(n)$ model with an interaction described by Eq. (6), in the limit $n\rightarrow\infty$, is given by the expression

$$\begin{aligned} \beta f(K,h|\mathbf{L},\mathbf{\Lambda}) = \frac{1}{2}\sup_{\tau>0}\Bigg\{ -\frac{h^2}{K\tau} + \frac{1}{L_1\times\cdots\times L_d} \\ \times \sum_{\mathbf{k}} \ln[\tau+\omega(\mathbf{k})] - K\tau \Bigg\} + \frac{1}{2}\left[\ln\frac{K}{2\pi} - \frac{K}{v_2}\right]. \end{aligned} \tag{34}$$

In the presence a sharp cutoff in k space, this leads to

$$\Delta U_{d,\sigma}^{(1)}(\tau,L,\Lambda) = \frac{\Lambda^d}{L^2}\frac{1}{6}\frac{1}{(2\pi)^{d-2}}\int_{-1}^{1}dm_2\cdots\int_{-1}^{1}dm_d \frac{1-\frac{1}{2}b\sigma\Lambda^{\sigma-2}(1+\theta^2)^{\sigma/2-1}+2c^2\Lambda^2(1+\theta^2)}{\tau+\Lambda^2(1+\theta^2)-b\Lambda^\sigma(1+\theta^2)^{\sigma/2}+2c^2\Lambda^4(1+\theta^2)^2}, \tag{35}$$

where $\theta^2 = m_2^2 + \cdots + m_d^2$. This term is missing in Eq. (9) in Ref. [18], but it is manifestly present in the case of a sharp cutoff. We therefore conclude that Eq. (9) of Ref. [18] is only correct within the smooth cutoff procedure, but not within the sharp cutoff one. As already explained above, Eq. (9) of Ref. [18] coincides with our Eq. (32) for systems with $\eta=0$ and $d+\sigma<6$ for *periodic* boundary conditions. In Ref. [18], it is supposed to be valid also for systems with *Dirichlet* boundary conditions. However, Dirichlet boundary conditions are inconsistent with the long-ranged nature of the dispersion forces (subleading long-ranged interaction) because the "missing neighbors" of the ordering degrees of freedom at a surface of such a system by the nature of long-ranged interactions generate a long-ranged surface field. We therefore conclude that the consideration of systems with subleading long-range interactions combined with Dirichlet boundary conditions as proposed in Ref. [18] is of no physical relevance. In the following section, we summarize our findings and also comment on the proper boundary conditions and the expected finite-size behavior of systems with dispersion forces and real boundaries. We will present arguments as to why we expect this to differ significantly from Eq. (32).

IV. SUMMARY AND CONCLUDING REMARKS

It has been shown in Secs. II and III that nonanalyticities in the dispersion relation $\omega(\mathbf{k})$ at the momentum cutoff lead to a bulk model which has leading and competing long-ranged interactions in real space. Thus even in the bulk it has peculiar properties such as the two-point correlation function given by Eq. (10). For such a model, finite-size scaling developed for the systems with short-ranged or subleading long-ranged interactions does not apply from the outset. We are not aware of any physical system governed by such a type of interaction.

In order to investigate the critical behavior of a model system by means of an effective Ginzburg-Landau Hamiltonian in $\mathbf{k}$ space, one usually expands $\omega(\mathbf{k})$ in the (infrared) limit $k\rightarrow 0$, keeping only the leading term(s). Divergent momentum integrals can be regularized by the introduction of a cutoff which is motivated by the presence of a Brillouin zone (lattice models) or finite particle sizes (continuum models). If a sharp cutoff in the momentum space is applied as a *strict* feature of an otherwise approximate (expanded) dispersion relation, the ensuing nonanalyticity of $\omega(\mathbf{k})$ at the cutoff generates long-ranged correlations and finite-size effects of the order L^{-2}. These effects do not occur in actual systems with short-ranged or subleading long-ranged interactions and therefore they are artificial. In particular, the nonuniversal long-ranged Casimir forces reported in Ref. [18] have no physical relevance for systems with short-ranged forces.

It is instructive to consider the finite-size behavior of the free energy in the systems with subleading long-ranged interactions. As is well known, the free energy decomposes into a sum of a regular and a singular part. In Sec. III we have discussed the finite-size behavior of the singular part for the case of periodic boundary conditions in a film geometry. However, the regular part is also important and it has experimental consequences for the Casimir force. For periodic boundary conditions, one expects the regular part to be of the order L^{-4} in the vicinity of T_c. Much more interesting is the case of a system with real boundaries. This raises the question of proper boundary conditions for such systems. The boundary conditions cannot be of the Dirichlet or Neumann type, because the latter are incompatible with the long-ranged nature of the interactions. Instead, one finds that the long-ranged interactions generate long-ranged *surface fields* that decay according to a power law away from the surface into the bulk of the system. The direct (Hamaker) interaction of the surfaces then generates a $L^{-\sigma}$ contribution to the regular part of the free energy if the free energy is measured per unit volume (or a $L^{-\sigma+1}$ contribution if the free energy is measured per unit area). This is well known from studies of wetting phenomena [17]. The contribution to the regular part of the free energy due to the action of the surface fields on the ordering degrees of freedom is also of the order of $L^{-\sigma}$. The available renormalization group arguments suggest that the contribution to the singular part should be of the order $L^{-(d-2+\eta)/2-\sigma}$ [61,62]. This leads to the following *hypothesis for the singular part of the free energy*:

$$f_s(t,L)=L^{-d}X(L/\xi,h_1L^{\Delta_1/\nu},bL^{2-\sigma-\eta},h_sL^{(d+2-\eta)/2-\sigma}), \tag{36}$$

where h_s is a nonuniversal metric factor characterizing the long-ranged behavior of the surface fields, while h_1 is the corresponding factor at the surface boundaries. Here Δ_1 is the critical surface gap exponent of the corresponding surface universality class. Note that, since $d>2$, the term proportional to h_s can give contributions that are *larger* than that proportional to b, i.e., the contributions of the surface fields are important and cannot be ignored. In particular, this is important for the quantitave analysis of wetting experiments with critical binary liquid mixtures [50]. The interpretation of these experimental data [50] is still unresolved. We also point out that neither Eq. (32) nor Eq. (36) applies to confined superfluid ^{4}He or ^{3}He-^{4}He mixtures [48], which have been under investigation in recent experiments [51,52]. For superfluid ^{4}He, the long-ranged finite-size contributions to the free energy originate from two distinct sources: (i) a *regular* contribution from the dispersion forces which couple to the fluid *density* and are unrelated to the superfluid order parameter; (ii) a *singular* contribution from the superfluid order parameter which is generically short ranged in nature and does not relate to the presence of dispersion forces [48]. The situation is more complicated in the case of ^{3}He-^{4}He mixtures, where the superfluid transition temperature depends on the ^{3}He concentration, which eventually causes the superfluid transition to become first order beyond the tricritical ^{3}He concentration. The ^{3}He concentration does respond to the dispersion forces and, by virtue of long-ranged surface fields, long-ranged ^{3}He concentration perturbations may emerge in a confined ^{3}He-^{4}He mixture. Through the dependence of the superfluid transition temperature T_λ on the ^{3}He concentration, a long-ranged variation of the local value of T_λ will ensue, which in turn imposes a corresponding variation of the superfluid density in thermal equilibrium. From the experiment in Ref. [52], there is robust evidence that the Dirichlet boundary conditions do not apply for the superfluid order parameter of ^{3}He-^{4}He in the tricritical regime, because contrary to the case of pure ^{4}He [51] a thickening of the wetting layer has been observed caused by a *repulsive* tricritical Casimir force. This observation rules out the pure Dirichlet boundary conditions in this case because these can only account for *attractive* Casimir forces as observed in pure ^{4}He [48,51]. Furthermore, subdominant long-ranged interactions may play a significant role in a finite-size scaling analysis of ^{3}He-^{4}He mixtures in the vicinity of the bulk superfluid transition. However, the model Hamiltonian of the system then has to accomodate a second "noncritical" field (the ^{3}He concentration) apart from the superfluid order parameter in order to include dispersion forces and long-ranged surface fields in the physically correct way. The construction of a proper model Hamiltonian, which is a generalization of the standard Ginzburg-Landau Hamiltonian considered here, is beyond the scope of this work.

In conclusion, we remark that we still lack a complete theoretical description of the Casimir effect in the systems with subleading long-ranged interactions. Such a description must contain both the influence of surface fields and the long-ranged nature of the interaction potential. Both are expected to generate important contributions to the critical behavior pertaining to the universality class of systems with short-ranged interaction potentials. Very close to T_c the finite-size behavior should turn out to be that of short-ranged models, but additional finite-size effects are expected to become dominant for $|t|L^{1/\nu} \gg 1$. For systems with scalar order parameter, these expectations hold both above and below the bulk critical temperature. For $O(n)$ models one expects that Goldstone modes will dominate the finite-size behavior of the systems below T_c.

ACKNOWLEDGMENTS

The authors thank Professor K. Binder, Professor R. Evans, Professor M. E. Fisher, and Professor J. Rudnick for a critical reading of the manuscript. D. Dantchev acknowledges the hospitality of Max-Planck-Institute for Metals Research in Stuttgart as well as the financial support of the Alexander von Humboldt Foundation.

[1] H.B.G. Casimir, Proc. K. Ned. Akad. Wet. **51**, 793 (1948).
[2] H.B.G. Casimir, Physica (Amsterdam) **19**, 846 (1953).
[3] V.M. Mostepanenko and N.N. Trunov, *The Casimir Effect and its Applications* (Oxford University Press, New York, 1997).
[4] K.A. Milton, *The Casimir Effect* (World Scientific, Singapore, 2001).
[5] M.E. Fisher and P.G. de Gennes, C. R. Seances Acad. Sci., Ser. B **287**, 207 (1978).
[6] M. Krech, *The Casimir Effect in Critical Systems* (World Scientific, Singapore, 1994).
[7] J.G. Brankov, D.M. Danchev, and N.S. Tonchev, *The Theory of Critical Phenomena in Finite-Size—Systems-Scaling and Quantum Effects* (World Scientific, Singapore, 2000).
[8] B.W. Harris, F. Chen, and U. Mohideen, Phys. Rev. A **62**, 052109 (2000).
[9] U. Mohideen and A. Roy, Phys. Rev. Lett. **81**, 4549 (1998).
[10] S.K. Lamoreaux, Phys. Rev. Lett. **78**, 5 (1997).
[11] G. Bressi, G. Carugno, R. Onofrio, and G. Ruoso, Phys. Rev. Lett. **88**, 041804 (2002).
[12] A. Lambrecht, Phys. World **15**(9), 29 (2002).
[13] R. Evans and J. Stecki, Phys. Rev. B **49**, 8842 (1994).
[14] A.O. Parry and R. Evans, Physica A **181**, 250 (1992).
[15] R. Evans, U. Marini Bettolo Marconi, and P. Tarazona, J. Chem. Phys. **84**, 2376 (1986).
[16] U. Marini Bettolo Marconi, Phys. Rev. A **38**, 6267 (1988).
[17] S. Dietrich, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, London, 1988), Vol. 12, p. 1.
[18] X.S. Chen and V. Dohm, Phys. Rev. E **66**, 016102 (2002).
[19] J. Zinn-Justin, *Quantum Field Theory and Critical Phenomena* (Clarendon, Oxford, 2002).
[20] D.J. Amit, *Field Theory, the Renormalization Group, and Critical Phenomena* (World Scientific, Singapore, 1984).
[21] M.E. Fisher, J. Math. Phys. **6**, 1643 (1965).
[22] M.E. Fisher, in *Critical Phenomena*, Proceedings of the 1970 International School of Physics ''Enrico Fermi,'' Course LI, edited by M. S. Green (Academic, New York, 1971), p. 1.
[23] V. Privman, P.C. Hohenberg, and A. Aharony, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J.L. Lebowitz (Academic, London, 1991), Vol. 14, p. 1.
[24] M.E. Fisher, S.-K. Ma, and B.G. Nickel, Phys. Rev. Lett. **29**, 917 (1972).
[25] M. Aizenman and R. Fernández, Lett. Math. Phys. **16**, 39 (1988).
[26] E. Brézin, J. Zinn-Justin, and J.C. Le Guillou, J. Math. Phys. **9**, L119 (1976).
[27] J. Frölich and T. Spencer, Commun. Math. Phys. **84**, 87 (1982).
[28] J. Cardy, *Scaling and Renormalization in Statistical Physics* (Cambridge University Press, Cambridge, 1996).
[29] J. Sak, Phys. Rev. B **8**, 281 (1973).
[30] J. Honkonen and M.Yu. Nalimov, J. Phys. A **22**, 751 (1989).
[31] J. Honkonen, J. Phys. A **23**, 825 (1990).
[32] H.K. Janssen, Phys. Rev. E **58**, R2673 (1998).
[33] E. Luijten and H.W.J. Blöte, Phys. Rev. Lett. **89**, 025703 (2002).
[34] R.J.F. Leote de Carvalho, R. Evans, D.C. Hoyle, and J.R. Henderson, J. Phys.: Condens. Matter **6**, 9275 (1994).
[35] R.F. Kayser and H.J. Raveché, Phys. Rev. A **29**, 1013 (1984).
[36] G. Flöter and S. Dietrich, Z. Phys. B: Condens. Matter **97**, 213 (1995).
[37] D.M. Dantchev, Eur. Phys. J. B **23**, 211 (2001).
[38] B.M. McCoy and T.T. Wu, *The Two-dimensional Ising Model* (Harvard University Press, Cambridge, 1973).
[39] M.E. Fisher and R.J. Burford, Phys. Rev. **156**, 583 (1967).
[40] X.S. Chen and V. Dohm, Eur. Phys. J. B **15**, 283 (2000).
[41] M.E. Fisher, in *Critical Phenomena*, Proceedings of 51st Enrico Fermi Summer School, Varenna, edited by M.S. Green (Academic, New York, 1972), p. 1.
[42] M.E. Fisher and M. Barber, Phys. Rev. Lett. **28**, 1516 (1972).
[43] M. Barber, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J.L. Lebowitz (Academic Press, New York, 1983), Vol. 8, p. 145.
[44] V. Privman and M.E. Fisher, Phys. Rev. B **30**, 322 (1984).
[45] V. Privman, in *Finite Size Scaling and Numerical Simulation of Statistical Systems*, edited by V. Privman (World Scientific, Singapore, 1990), p. 1.
[46] D. Dantchev and J. Rudnick, Eur. Phys. J. B **21**, 251 (2001).
[47] Here $\xi \equiv \xi(L,t)$ is the finite-size correlation length, see e.g., Ref. [7]; $\xi(L \to \infty, t) \to \xi(\infty, t)$, with $\xi(\infty,t) \simeq \xi_0^+ t^{-\nu}$, where $\nu = 1/(d-2)$.
[48] M. Krech and S. Dietrich, Phys. Rev. Lett. **66**, 345 (1991); Phys. Rev. A **46**, 1886 (1992); **46**, 1922 (1992).
[49] M. Krech, J. Phys.: Condens. Matter **11**, R391 (1999).
[50] A. Mukhopadhyay and B.M. Law, Phys. Rev. Lett. **83**, 772 (1999); Phys. Rev. E **62**, 5201 (2000).
[51] R. Garcia and M.H.W. Chan, Phys. Rev. Lett. **83**, 1187 (1999).
[52] R. Garcia and M.H.W. Chan, Phys. Rev. Lett. **88**, 086101 (2002).
[53] X.S. Chen and V. Dohm, Eur. Phys. J. B **7**, 183 (1999).
[54] X.S. Chen and V. Dohm, Eur. Phys. J. B **10**, 687 (1999).
[55] J.L. Cardy, in *Fields, Strings and Critical Phenomena*, edited

by E. Brézin and J. Zinn-Justin (Elsevier, Amsterdam, 1989), p. 169.
[56] X.S. Chen and V. Dohm, e-print cond-mat/0108202.
[57] G. Parisi, *Statistical Field Theory* (Addison-Wesley, Reading, MA, 1992).
[58] See ''Note added in proof,'' p. 700, of Ref. [54].
[59] H. Chamati and D.M. Dantchev, Eur. Phys. J. B **26**, 89 (2002).
[60] V. Privman and M.E. Fisher, Phys. Rev. B **30**, 322 (1984).
[61] L. Peliti and S. Leibler, J. Phys. C **16**, 2635 (1983).
[62] H.W. Diehl, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J.L. Lebowitz (Academic, London, 1986), Vol. 10, p. 76.