Violation of finite-size scaling for the free energy near criticality

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The singular part of the finite-size free energy density $f_s$ of the $O(n)$ symmetric $\varphi^4$ field theory is calculated for confined geometries of linear size $L$ with periodic boundary conditions in the large-$n$ limit and with Dirichlet boundary conditions in one-loop order. We find that both a sharp cutoff and a subleading long-range interaction cause a non-universal $L$ dependence of $f_s$ near $T_c$. For film geometry this implies a non-universal critical Casimir force with an algebraic $L$ dependence that dominates the exponential finite-size scaling behavior above $T_c$ for both periodic and Dirichlet boundary conditions.

Near bulk criticality Eqs. (1) - (5) yield

$$f_c(x, L) = L^{-d} X_{c}(L/\xi) \quad \text{(6)}$$

where $X_c(x) = (d-1)F(x) - x F'(x) + Y x^d \quad \text{(7)}$

with $F'(x) = \partial F(x)/\partial x$. The universal scaling structure of Eqs. (6) and (7) has been confirmed by renormalization-group (RG) and model calculations and is predicted to be valid also for Dirichlet boundary conditions (Dbc).

In this Letter we show that the scaling predictions of Eqs. (3) and (4) are significantly less generally valid than anticipated previously. On the basis of exact results for the $O(n)$ symmetric $\varphi^4$ field theory in the large-$n$ limit and on the basis of one-loop results we shall identify two sources in the $\varphi^4$ Hamiltonian that cause non-scaling finite-size effects: (i) a short-range interaction term $\sim k^2$ with a sharp cutoff $\Lambda$ in $k$ space, (ii) an additional subleading long-range interaction term $\sim b|k|^\sigma$, $2 < \sigma < 4$. In the latter case we consider both pbc and Dbc. Specifically, we find for $2 < d < 4$ that Eqs. (3) and (4) must be complemented as

$$f_s(t, L, \Lambda) = L^{-2} \Lambda^{-2} \Phi(\xi^{-1} \Lambda^{-1}) + L^{-d} F(L/\xi) \quad \text{(8)}$$

where $F(x)$ is a universal scaling function and $\xi(t)$ is the bulk correlation length. Both $\xi$ and $L$ are assumed to be sufficiently large compared to microscopic lengths (for example, the lattice spacing $\alpha$ of lattice models, the inverse cutoff $\Lambda^{-1}$ of field theories, or the length scale of subleading long-range interactions). Eq. (3) includes the bulk limit $f_s(t, \infty) = f_{bs}(t) = Y \xi^{-d}$ with a universal amplitude $Y$. Eqs. (1) - (3) are expected to remain valid also for non-cubic geometries where the scaling function $F(x)$ depends on the geometry and on the universality class of the bulk critical point but not on $\alpha$ or $\Lambda$ and not on other interaction details. In particular subleading long-range interactions (such as van der Waals forces in fluids) that do not affect the universal bulk critical behavior are assumed to yield only negligible finite-size corrections to $F(x)$.

As a consequence, universal finite-size scaling properties are generally believed to hold for observable quantities derived from $f_s(t, L)$, such as the critical Casimir force $F_C$ in film geometry

$$F_C = -\partial f_c^\infty(t, L)/\partial L \quad \text{(4)}$$

where the excess free energy per unit area is given by

$$f_c^\infty(t, L) = L f(t, L) - L f_0(t) \quad \text{(5)}$$

Near bulk criticality Eqs. (4) and (5) yield

$$F_C(\xi, L) = L^{-d} X_C(L/\xi) \quad \text{(6)}$$

where

$$X_C(x) = (d-1)F(x) - x F'(x) + Y x^d \quad \text{(7)}$$

The concept of universal finite-size scaling has played an important role in the investigation of finite-size effects near critical points over the last decades. Consider the free-energy density $f(t, L)$ of a finite system at the reduced temperature $t = (T - T_c)/T_c \geq 0$ and at vanishing external field in a $d$-dimensional cubic geometry of volume $L^d$ with periodic boundary conditions (pbc). It is well known that, for small $t$, the bulk free energy density $f_b(t) \equiv f(t, \infty)$ can be decomposed as

$$f_b(t) = f_{bs}(t) + f_0(t) \quad \text{(1)}$$

where $f_{bs}(t)$ denotes the singular part of $f_b$ and where the regular part $f_0(t)$ can be identified unambiguously. According to Privman and Fisher the singular part of the finite-size free-energy density may be defined by

$$f_s(t, L) = f(t, L) - f_0(t) \quad \text{(2)}$$

where $f_0$ is independent of $L$. The finite-size scaling hypothesis asserts that, below the upper critical dimension $d = 4$ and in the absence of long-range interactions, $f_s(t, L)$ has the structure

$$f_s(t, L) = L^{-d} F(L/\xi) \quad \text{(3)}$$

where $F(x)$ is a universal scaling function and $\xi(t)$ is the bulk correlation length. Both $\xi$ and $L$ are assumed to be sufficiently large compared to microscopic lengths (for example, the lattice spacing $\alpha$ of lattice models, the inverse cutoff $\Lambda^{-1}$ of field theories, or the length scale of subleading long-range interactions). Eqs. (3) includes the bulk limit $f_s(t, \infty) = f_{bs}(t) = Y \xi^{-d}$ with a universal amplitude $Y$. Eqs. (1) - (3) are expected to remain valid also for non-cubic geometries where the scaling function $F(x)$ depends on the geometry and on the universality class of the bulk critical point but not on $\alpha$ or $\Lambda$ and not on other interaction details. In particular subleading long-range interactions (such as van der Waals forces in fluids) that do not affect the universal bulk critical behavior are assumed to yield only negligible finite-size corrections to $F(x)$.

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The singular part of the finite-size free energy density $f_s$ of the $O(n)$ symmetric $\varphi^4$ field theory is calculated for confined geometries of linear size $L$ with periodic boundary conditions in the large-$n$ limit and with Dirichlet boundary conditions in one-loop order. We find that both a sharp cutoff and a subleading long-range interaction cause a non-universal $L$ dependence of $f_s$ near $T_c$. For film geometry this implies a non-universal critical Casimir force with an algebraic $L$ dependence that dominates the exponential finite-size scaling behavior above $T_c$ for both periodic and Dirichlet boundary conditions.
for the case (i), and
\[ F_C(\xi, L, b) = -b L^{-d+2-\sigma} B(L/\xi) + L^{-d} X_C(L/\xi) \] (9)
for the case (ii), where the function \( \Phi \) has a finite critical value \( \Phi(0) > 0 \) and where the function \( B(L/\xi) \) has a non-exponential decay \( \sim (L/\xi)^{-2} \) above \( T_c \) for both pbc and Dbc. This implies (i) that the non-scaling \( L^{-2} \) term in Eq. (8) exhibits a dominant size dependence compared to the \( L^{-d} \) scaling term and (ii) that the non-universal term proportional to \( b \) in Eq. (8) implies an algebraic \( L \) dependence \( \sim b \xi^2 L^{-d-\sigma} \) that dominates the exponential finite-size scaling term of \( X_C \) above \( T_c \) for both pbc and Dbc. By contrast, for the \( \varphi^4 \) lattice model with short-range interaction, we find that Eqs. (3) and (8) are indeed valid except that for \( L \gg \xi \) above \( T_c \) the exponential scaling arguments of \( F \) and \( X_C \) must be formulated in terms of the lattice-dependent “exponential” correlation length \( \xi_0 \).

Non-negligible cutoff effects \( \Xi_1 \) and non-universal finite-size effects due to subleading long-range interactions \( \Xi_2 \) were already found previously for the finite-size susceptibility. These effects, however, were restricted to the regime \( L \gg \xi \) close to the bulk limit above \( T_c \). The new non-scaling finite-size effect (i) exhibited in Eq. (8) is significantly more general in that it is pertinent to the entire \( \xi^{-1} - L^{-1} \) plane including the central finite-size regime \( \xi \gg L \). In particular, this effect exists at \( T_c \) where \( f_\infty(0, L, \Lambda) \) has a non-universal leading amplitude \( L^{-2} \xi^2 \phi(0) \) for \( d > 2 \). This implies the non-universality of the critical Casimir force \( L^{-2} \xi^2 \phi(0) \) in film geometry.

Furthermore, the new non-scaling effect (ii) exhibited in Eq. (8) has relevant physical consequences in systems with subleading long-range interactions. These consequences are significantly more important than those considered previously for the finite-size susceptibility for pbc \( \Xi_1 \) \( \Xi_2 \). The latter are of limited physical relevance since in real systems they are dominated by surface terms of \( O(L^{-1}) \). In this Letter we predict a leading non-scaling effect on \( F_C \) not only for model systems with pbc but also for real systems with Dbc.

We start from the standard \( \varphi^4 \) continuum Hamiltonian
\[ H = \int d^d x \left[ \frac{1}{2} r_0 \varphi^2 + \frac{1}{2} (\nabla \varphi)^2 + u_0 (\varphi^2)^2 \right] \] (10)
with \( r_0 = r_{0c} + a_0 \) for the \( n \)-component field \( \varphi(x) \) in a partially confined \( L^d \times \infty^{d-d'} \) geometry with periodic boundary conditions in \( d' < d \) dimensions. This model requires a specification of the \( x \) dependence of \( \varphi(x) \) at short distances. We decompose the vector \( x \) as \( (y, z) \) where \( z \) denotes the coordinates in the \( d' \) confined directions. We consider two cases (i) and (ii).

Case (i): We assume pbc and a sharp cutoff \( \Lambda \), i.e., we assume that the Fourier amplitudes \( \hat{\varphi}_{p,q} \) of \( \varphi(y, z) = L^{-d} \sum_p \sum_q \hat{\varphi}_{p,q} e^{i(p x + q y)} \) are restricted to wavevectors \( p \) and \( q \) with components \( p_j \) and \( q_j \) in the range \( -\Lambda \leq p_j < \Lambda \) and \( |q_j| \leq \Lambda \). Here \( \int_{\mathbf{q}} \) stands for \( (2\pi)^{-d-2} \int d^{d-d'} q \), and \( \sum_p \) runs over \( p_j = 2\pi m_j / L \) with \( m_j = 0, \pm 1, \pm 2, \ldots \). The question can be raised whether there exists a non-negligible cutoff dependence of the finite-size free energy density per component (divided by \( k_B T \)),
\[ f_{d,d'}(t, L, \Lambda) = -n^{-1} L^{-d} \lim_{L \to \infty} \bar{L}^{-d+d'} \ln Z_{d,d'} \] (11)
where
\[ Z_{d,d'}(t, L, \bar{L}, \Lambda) = \prod_{k,q} \int d \hat{\varphi}_{p,q} \frac{\hat{\varphi}_{p,q}}{\Lambda(d-2)/2} \exp(-H) \] (12)
is the dimensionless partition function of a \( L^d \times \bar{L}^{d-d'} \) geometry. For comparison we shall also consider the free energy density \( \tilde{f}(t, L, \bar{L}) \) of the \( \varphi^4 \) lattice model
\[ \tilde{H} = \tilde{a}^d \left[ \sum_i \left( \frac{1}{2} \varphi_i^2 + u_0 (\varphi_i^2)^2 \right) + \sum_{i<j} \frac{J}{2a^2} (\varphi_i - \varphi_j)^2 \right] \] (13)
with a nearest-neighbor coupling \( J \) on a simple-cubic lattice with a lattice spacing \( \tilde{a} \). The factor \( (k_B T)^{-1} \) is absorbed in \( H \) and \( \tilde{H} \).

We shall answer this question in the exactly solvable limit \( n \to \infty \) at fixed \( u_0 n \) where the free energy density is \( 20 \).
\[ f_{d,d'}(t, L, \Lambda) = -\frac{1}{2} \Lambda^d \ln \pi - \frac{(r_0 - \chi^{-1})^2}{16 u_0 n} \]
\[ + \frac{1}{2} L^{-d''} \sum_{k,q} \ln \left[ \Lambda^{-2}(\chi^{-1} + p^2 + q^2) \right] \] (14)
Here \( \chi^{-1} \) is determined implicitly by
\[ \chi^{-1} = r_0 + 4u_0 n L^{-d} \sum_{k,q} \left( \chi^{-1} + p^2 + q^2 \right)^{-1} \] (15)
The bulk free energy \( f_0 \) and bulk susceptibility \( \chi_b \) above \( T_c \) are obtained by the replacement \( L^{-d} \sum_p \sum_q \rightarrow \int_{\mathbf{k}} \), and the critical point is determined by \( r_0 = r_{0c} = -4u_0 \int_k k^2 \) where \( \mathbf{k} \equiv (p, q) \). The bulk correlation length above \( T_c \) is \( \xi = \chi_b^{1/2} = \xi_0 \nu \) where \( \nu = (d-2)^{-1} \). The regular part of \( f_0 \) reads \( f_0 = \tilde{c}_4 \Lambda^d - r_{0c}^2 / (16u_0 n) \) where \( \tilde{c}_4 \) is a \( d \) dependent constant. The singular part of \( f_0 \) above \( T_c \) is \( f_{bs} = Y \xi^{-d} \) with the universal amplitude \( Y = (d-2)A_d / [2d(4-d)] \) where \( A_d = 2^{2-d} \pi^{-d/2} (d-2)^{-1} \Gamma(3-d/2) \). For the singular part \( f_s = f_{d,d'} - f_0 \) of the finite-size free energy above and at \( T_c \) we find the form of Eq. (8) with the leading non-scaling part
\[ \Phi_{d,d'}(\xi^{-1} \Lambda^{-1}) = \frac{d'}{6(2\pi)^{d-2}} \int_0^\infty dy \left[ \int dq e^{-q^2 y} \right]^{d-1} \times \exp \left[ -(1 + \xi^{-2} \Lambda^{-2} y) \right] \] (16)
and the subleading universal scaling part

\[ F_{d,d'}(L/\xi) = \frac{A_d}{2(4-d)} \left( \frac{L/\xi}{d-2} P^2 - \frac{2}{d} P^d \right) \]

\[ + \frac{1}{2} \int_0^\infty \frac{dy}{y} \left( \sqrt{\frac{\pi}{y}} \right)^{d-2} W_d(y) e^{-P^2 y/4 \pi^2} \]  

(17)

where \( P(L/\xi) \) is determined implicitly by

\[ P^{d-2} = \left( \frac{L/\xi}{d-2} \right)^{d-2} - \frac{4 - d}{4 \pi^2 A_d} \int_0^\infty dy \left( \sqrt{\frac{\pi}{y}} \right)^{d-2} \times \]

\[ \times W_d(y) e^{-P^2 y/4 \pi^2} , \]  

(18)

\[ W_d(y) = \left( \sqrt{\frac{\pi}{y}} \right) - \left( \sum_{m=-\infty}^{\infty} e^{-y m^2} \right)^d . \]  

(19)

This result remains valid also for \( t < 0 \) after replacing the terms \( (L/\xi)^{d-2} \) in Eqs. (17) and (18) by \( t(L/\xi^0)^{d-2} \) and after dropping the term \( -\xi^{-2} L^{-2} y \) in the exponent of Eq. (16). We have confirmed the structure of Eq. (8) also for the \( \varphi^4 \) theory with finite \( n \) within a one-loop RG calculation at finite \( \Lambda \) which yields the same form of the function \( \Phi_{d,d'}(\xi^{-1} L^{-1}) \) as in Eq. (14). Thus the universal scaling form (8) is invalid for the \( \varphi^4 \) field theory with pbc and with a sharp cutoff, both for \( T \geq T_c \) and for \( T < T_c \).

These results have a significant consequence for the critical Casimir effect. Instead of Eq. (12) we obtain from Eqs. (8) and (13) - (14) in film geometry \( (d' = 1) \)

\[ F_{C}(\xi, L, \Lambda) = L^{-2} \Lambda^{-2} \Phi_{d,1} (\xi^{-1} \Lambda^{-1}) + L^{-d} X_C(L/\xi) . \]  

(20)

Thus the critical Casimir force has a leading non-universal term \( \sim L^{-2} \), in addition to the subleading universal terms \( \sim L^{-d} \) of previous theories [11,12], both for \( T \geq T_c \) and for \( T < T_c \).

We have also calculated \( f_{d,d'} \) and \( F_{C} \) for the lattice Hamiltonian [13] and for the continuum Hamiltonian [14] with a smooth cutoff in the large-n limit. In both cases the scaling form (8) is found to be valid. For the lattice model, however, the second-moment bulk correlation length \( \xi \) in the argument of \( \Phi \) must be replaced by the lattice-dependent exponential correlation length [13,14]. Specifically we find, at fixed \( t > 0 \), the exponential large-\( L \) behavior

\[ f_s(t, L, \tilde{a}) - f_{bs} = -d'(L/2\pi \xi_1)^{(d-1)/2} L^{-d} \exp(-L/\xi_1) \]

(21)

where \( \xi_1 = (\tilde{a}/2) [\arcsinh(\tilde{a}/2\xi)]^{-1} \) is the exponential correlation length in the direction of one of the cubic axes. Note that the non-universal dependence of \( \xi_1 \) on \( \tilde{a} \) is non-negligible in the exponent of [21, 23].

The sensitivity of \( f_s(t, L, \Lambda) \) and \( F_{C} \) with respect to the cutoff procedure can be explained in terms of a corresponding sensitivity of the bulk correlation function

\[ G(x) = < \varphi(x)\varphi(0) > \]  

in the range \(|x| \gg \xi |13| \). For example, for the \( \varphi^4 \) continuum Hamiltonian [11] with an isotropic sharp cutoff \(|k| \leq \Lambda \) we find, in the large-\( n \) limit, the oscillatory power-law decay above \( T_c \)

\[ G(x) = 2 \Lambda^{-2} \left( 2\pi x \Lambda \right)^{-\sigma(d+1)/2} \sin \left( \frac{\pi x - \pi(d-1)/4}{1 + \xi^{-2} \Lambda^{-2}} \right) \]

(22)

for large \( x = |x| \gg \xi \) corresponding to the existence of long-range spatial correlations which dominate the exponential scaling dependence \( \sim e^{-x/\xi} \). By contrast, \( G(x) \) has an exponential decay for the lattice model [14] with purely short-range interaction [15]. An exponential decay of \( G(x) \) is also valid for the continuum model (10) with a smooth cutoff [13].

The non-universal cutoff effects on \( f_s \), \( F_{C} \) and on \( G(x) \) described above are a consequence of the long-range correlations induced by the sharp-cutoff procedure in the presence of pbc. We consider these consequences not only as a mathematical artifact but rather as an important signal for a serious lack of universality in physical systems. We substantiate this interpretation by demonstrating that corresponding violations of finite-size scaling should indeed exist in physical systems with more realistic interactions and boundary conditions.

Case (ii) : We assume the existence of a subleading long-range interaction in the continuum \( \varphi^4 \) Hamiltonian \( H \) which in the Fourier representation has the form \( b|k|^\sigma \) with \( 2 < \sigma < 4 \), in addition to the short-range term \( k^2 \). It is well known that the subleading interaction \( \sim |k|^\sigma \) corresponds to a spatial interaction potential \( V(x) \sim |x|^{-d-\sigma} \) that does not change the universal bulk critical behavior [21]. Interactions of this type exist in real fluids. As pointed out by Dantchev and Rudnick [14], the presence of this interaction yields leading non-scaling finite-size effects on the susceptibility \( \chi \) for the case of pbc in the regime \( L \gg \xi \) above \( T_c \), similar to those found for a sharp cutoff [11,15].

In real systems with non-periodic boundary conditions, however, these non-scaling finite-size effects become only subleading correlations that are dominated by the surface terms of \( \chi \) of \( O(L^{-1}) \). In the following we show that the situation is fundamentally different for \( F_{C} \) which, by definition, does not contain contributions of \( O(L^{-1}) \) arising from the \( O(L^{-1}) \) part \( \tilde{f} \) of the free energy density.

We consider film geometry and first assume Dbc in the \( z \) direction corresponding to \( \varphi(y, z) = \varphi(y, L) = 0 \), i.e., we assume that \( \Sigma_p \), in the Fourier representation of \( \varphi(y, z) = L^{-1} \sum_p \tilde{\varphi}_{p, n} e^{ipy} \sin(pz) \) runs over \( p = \pi m/L, m = 1, 2, ... \) Such boundary conditions are relevant for the superfluid transition of \(^4\text{He} \) [22].
presence of the subleading interaction $b|k|^\sigma$ implies, for $L \gg \xi$ above $T_c$, a non-universal term $\sim b$ in

$$f_s(t, L, b) - \tilde{f} = -bL^{-d+2-\sigma} \Psi(L/\xi) + L^{-d} G(L/\xi)$$

(23)

where $G(L/\xi)$ is the known universal scaling function for purely short-range interaction. The scaling function $G(L/\xi)$ has an exponential large-$L$ behavior. By contrast we find that $\Psi(L/\xi)$ has an algebraic $L$ dependence.

Performing a one-loop RG calculation we obtain

$$\Psi(L/\xi) = \frac{1}{2} (2\pi)^{4-d} \int_0^\infty dx \left( 1 + 2 \frac{\partial}{\partial x} \right) \tilde{\Psi}(x),$$

(24)

$$\tilde{\Psi}(x) = \int_0^\infty dy y^{(2-\sigma)/2} e^{-xy/4\pi^2} \left( \sqrt{\pi} / y \right)^{d-1} \tilde{W}_1(y) \times$$

$$\times \gamma^* \left( \frac{2-\sigma}{2}, \frac{xy}{4\pi^2} \right)$$

(25)

where $\gamma^* (z, x) = x^{-z} \int_0^x dt e^{-t} t^{z-1} / \int_0^\infty dt e^{-t} t^{z-1}$ is the incomplete Gamma function and

$$\tilde{W}_1(y) = \sqrt{\pi} y^{-1} - \frac{1}{2} \sum_{n = -\infty}^\infty \exp \left( -\frac{y}{4} n^2 \right).$$

(26)

We have found that cutoff effects are negligible for the function $\Psi(L/\xi)$. At fixed $\xi$, the large-$L$ behavior is

$$\Psi(L/\xi) \sim (L/\xi)^{-2},$$

Eq. (23) yields the following form

$$B(L/\xi) = (d-3+\sigma) \Psi(L/\xi) - (L/\xi) \Psi'(L/\xi)$$

(27)

for the non-universal contribution to $F_C$ in Eq. (3). The crucial consequence is that the leading critical temperature dependence $\sim bL^{d-\sigma}$ of $F_C$ for $L \gg \xi$ above $T_c$ is algebraic and non-universal whereas the critical temperature dependence of the scaling part $X_C(L/\xi)$ derived from $G(L/\xi)$ is exponential and universal. This may have significant consequences for the interpretation of existing and future experimental data in fluids.

For comparison we finally present our result for $\Psi(L/\xi)$ for film geometry in the presence of pbc. In one-loop order we obtain for $\Psi_{pc} (L/\xi)$ the same form as given for $\Psi(L/\xi)$ in Eqs. (24) and (25) but with $\tilde{W}_1(y)$ replaced by $\tilde{W}_1(y)$, Eq. (43). For the large-$L$ behavior we find $\Psi_{pc} (L/\xi) \sim (L/\xi)^{-2}$ which dominates the exponential scaling dependence of $X_C$. Our prediction of a non-exponential non-scaling effect on $F_C$ above $T_C$ for pbc can be tested by Monte Carlo simulations.

The next stage of the theory would entail a quantitative determination of the non-universal interaction parameter $b$ for a specific system. This is, of course, beyond the scope of the present paper.

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