

# Off-Critical Casimir Effect in Ising Slabs with Symmetric Boundary Conditions in $d = 3$

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Extended de Gennes-Fisher (EdGF) local-functional method has been applied to the thermodynamic Casimir effect *away* from the critical point for systems in the Ising universality class confined between parallel plane plates with symmetric boundary conditions (denoted  $(ab) = (++)$ ). Results on the universal scaling functions of the Casimir force  $W_{++}(y)$  ( $y$  is a temperature-dependent scaling variable) and Gibbs adsorption  $\tilde{G}(y)$  are presented in spatial dimension  $d = 3$ . Also, the mean-field form of the universal scaling function of the Gibbs adsorption  $\tilde{G}(y)$  is derived within the local functional theory. Asymptotic behavior of  $W_{++}(y)$  for large values of the scaling variable  $y$  is analyzed in *general* dimension  $d$ .

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The Casimir effect in quantum or statistical physics refers to long-range forces that emerge due to a confinement on fluctuations. In statistical physics these fluctuations are in the order parameter of a thermodynamic system *at* or *near* the critical point, as predicted in 1978 [1]. The Casimir force (CF) depends on the nature of the confined system as well as the boundary conditions (b.c.) and the geometrical form of the confinement [2, 3]. Much theoretical work has examined various surface universality classes for Ising systems and classical fluids either exactly *at* criticality or *away* from it [3]. Symmetry breaking b.c. (defined below) are of particular interest for experiments with critical binary liquid mixtures. Appreciable agreement pertaining to these systems has been achieved between theory [3–5] and recent experiments on the CF *at* criticality in complete wetting films of binary/fluorocarbon mixture near liquid vapor coexistence [6], with the mean value of the universal Casimir amplitude (a measure of CF at the bulk critical point, defined below) most closely corresponding to earlier prediction of the local-functional theory [4], while at the same time encompassing other theoretical/simulation estimates [5]. Exact results on the full-temperature dependence of the CF are available in spatial dimensions  $d = 2$  [7] and  $d \geq 4$  (mean field theory) [5, 8] for both cases of symmetry breaking b.c.

Although significant theoretical effort has focused on the universal scaling functions of the off-critical Casimir effect, knowledge of them is still somewhat incomplete for spatial dimension  $d = 3$ , even for the relevant Ising universality class. Pertinent results in this case refer to films with periodic b.c., studied via Monte Carlo (MC) simulations [9] or by the field-theoretic approach for Dirichlet, Neumann and periodic b.c. [10], besides recent MC results that now include symmetry breaking b.c. [11] and are most relevant for the present content. In this Letter a thermodynamic system in the Ising universality class is considered in the vicinity of the bulk critical point. The system is confined between two parallel plane plates of

area  $A$  separated by distance  $L$ . We shall consider only those slabs where an external symmetry-breaking boundary field has been applied to both plates, i.e. a field  $h_1$  (respectively,  $h_2$ ) acting on the plate at  $z = 0$  (respectively,  $z = L$ ), and assume that fields  $h_1$  and  $h_2$  are of the same sign,  $h_1 h_2 > 0$ , corresponding to the so called symmetric b.c.

Thermodynamic CF is defined as a generalized force conjugate to separation  $L$  between the plates  $F_{\text{Casimir}}(T; L) := -\frac{\partial f^\times}{\partial L}$ , where  $f^\times(T; L)$  is the reduced incremental free energy defined by  $f^\times(T; L) := \lim_{A \rightarrow \infty} \frac{F}{k_B T_c A} - L f_b$  for free energy  $F$  with  $f_b$  being the reduced bulk free energy. It is characterized by the property  $F_{\text{Casimir}}(T; L) \rightarrow 0$  as  $L \rightarrow \infty$ . According to the finite-size scaling theory, critical phenomena near the bulk critical temperature  $T_c$  and bulk field  $h = 0$  are governed by universal scaling functions that depend on the ratio  $L/\xi$  [12–16], where  $\xi$  is the bulk correlation length with  $\xi(t, h = 0) \approx \xi_0^\pm |t|^{-\nu}$ , as the reduced temperature  $t = (T - T_c)/T_c \rightarrow 0^\pm$ ,  $\xi_0^\pm$  nonuniversal amplitudes and  $\nu$  a critical exponent. Then the CF can be expressed in terms of the universal scaling function  $W_{ab}(\cdot)$  [3]:

$$F_{\text{Casimir}}(T; L) = L^{-d^*} W_{ab}(y), \quad y = c_1 t L^{1/\nu}, \quad (1)$$

where  $c_1$  is a nonuniversal metric factor. The scaling function  $W_{ab}(y)$ , having universal shape [3], does depend on the definition of the correlation length. In order to allow for the “natural” scaling variable  $L/\xi$  ( $\xi$  is chosen as true correlation length) to emerge in the local-functional expressions of  $W_{ab}(y)$  in the asymptotic limits  $y \rightarrow \pm\infty$ , considered shortly, we choose  $c_1 = 1/(\xi_0^+)^{1/\nu}$ . Exactly at the critical temperature  $T_c$ , the scaling functions  $W_{ab}(\cdot)$  give the universal Casimir amplitudes [1, 3]  $A_{ab}$  via  $W_{ab}(0) = (d^* - 1)A_{ab}$ , as already considered within local-functional theory for symmetric and antisymmetric  $(+-)$  ( $h_1 h_2 < 0$ ) b.c. [4]. Note that  $d^* = \min(d, d_>)$ , where  $d$  is a spatial dimension and  $d_>$  the upper critical dimension of the system, the Ising universality class has  $d_> = 4$ .

The purpose of this Letter is to apply EdGF method introduced by Fisher and Upton [18], in order to examine the Casimir effect for systems of the Ising universality class under the symmetric (++) b.c. over the whole temperature range, in particular, in  $d = 3$ , important in respect to the experiments where more accurate theoretical analysis of the above quantities has been missing until now. As a nonperturbative approach, EdGF theory allows for calculation directly at a fixed spatial dimension, an advantage over field-theoretic approach in terms of  $\epsilon$  expansion.

The local-functional method [18] asserts that magnetization profile  $m(z)$  in film geometry is given by minimizing a (local) interfacial functional  $\mathcal{F}[m]$ :

$$\mathcal{F}[m] := \int_0^L \mathcal{A}(m, \dot{m}, t, h) dz + f_1(m_1; h_1) + f_2(m_2; h_2) \quad (2)$$

where  $m_1 = m(z = 0)$ ,  $m_2 = m(z = L)$  with  $f_i = -h_i m_i - g m_i^2/2$  ( $i = 1, 2$ ), the usual surface terms which allow for the presence of external walls (at  $z = 0$  and  $z = L$ ), and  $\dot{m} = dm/dz$ . The integrand  $\mathcal{A}$  is assumed to take the form which contains *only* bulk quantities [18]:  $\mathcal{A}(m, \dot{m}; t, h) = \{J(m)\mathcal{G}[\Lambda(m, t, h)\dot{m}] + 1\}W(m, t, h)$ , where  $W(m, t, h) = \Phi(m, t) - \Phi(m_b, t) - h(m - m_b)$ , and  $\Phi(m, t)$  is the bulk Helmholtz free energy density. The bulk magnetization is denoted by  $m_b$ , where for  $h = 0$ ,  $m_b = B(-t)^\beta$  for  $t < 0$  and  $m_b = 0$  for  $t > 0$  with  $\beta$  a critical exponent and  $B$  a nonuniversal amplitude. The function  $\mathcal{G}(x)$  is required to satisfy several properties [4, 18]. As before [18], we choose  $J(m) = 1$  and  $\Lambda(m; t, h) := \xi(m; t)/\sqrt{2\chi(m; t)W(m; t, h)}$ , where  $\xi(m; t)$  and  $\chi(m; t)$  are, respectively, the bulk correlation length and susceptibility of a homogenous system at  $(m, t)$ . Mean-field theory ( $d > 4$ ) follows from having  $\Phi(m; t)$  take Landau form with  $(\xi^2/2\chi)(m; t)$  being *constant* in  $m$  and  $t$ . For more general  $d > 1$ , bulk functions have the following analytic scaling forms [18]:

$$W(m; t, 0) \approx |m|^{\delta+1} Y_{\pm}(m/m_0(t)), \quad (3a)$$

$$(\xi^2/2\chi)(m; t) \approx |m|^{-\eta\nu/\beta} Z_{\pm}(m/m_0(t)), \quad (3b)$$

in the simultaneous scaling limits  $t \rightarrow 0^{\pm}$  and  $m \rightarrow 0$ , where  $m_0(t) := B|t|^\beta$ ,  $\eta$  is the critical bulk correlation function exponent in standard notation.

Minimization of the functional, Eq. (2), yields the magnetization profile,  $m(z)$ , which for  $h_1 > 0$ , contains a minimum at  $z = z_+$  with magnetization  $m_+ := m(z_+) := m_0(t)w$ . The scaling variable  $y$  and  $w$  are related solely in terms of universal quantities:

$$A_2 |y|^\nu = \int_w^\infty \frac{\sqrt{\tilde{Z}_{\pm}(u)/\tilde{Y}_{\pm}(u)} du}{u^{1+\nu/\beta} |\hat{\mathcal{G}}^{-1}[1 - (w/u)^{1+\delta} \tilde{Y}_{\pm}(w)/\tilde{Y}_{\pm}(u)]|} \quad (4)$$

where  $\hat{\mathcal{G}}(x) := x d\mathcal{G}/dx - \mathcal{G}$ ,  $A_2 := R_\chi \delta / [Q_2 \sqrt{2\delta(\delta+1)}]$  is defined by the standard universal amplitudes [19]  $R_\chi = C_+ B^{\delta-1} D$ ,  $Q_2 = (C_+/C_c)(\xi_c/\xi_0^+)^{2-\eta}$ ,  $C_+$  a nonuniversal zero-field susceptibility amplitude above the critical

temperature,  $D$ ,  $C_c$  and  $\xi_c$  defined along the critical isotherm: ( $T = T_c$ ),  $h \approx Dm^\delta$ , with  $C_c$  being the corresponding susceptibility amplitude  $\chi \approx C_c |h|^{-(1-1/\delta)}$  and  $\xi_c$  is defined from  $\xi(m; 0) \approx \xi_c |h|^{-\nu/\beta\delta}$ . The universal functions  $\tilde{Y}(\cdot)$  and  $\tilde{Z}(\cdot)$  are obtained from normalizing  $Y(\cdot)$  and  $Z(\cdot)$ , respectively. The local-functional calculation of the CF then follows from  $\partial f^\times/\partial L = W(m_+)$ , which yields the universal scaling function  $W_{++}(y)$  for  $d < 4$ :

$$W_{++}(y) = -A_1 |y|^{2-\alpha} w^{1+\delta} \tilde{Y}_{\pm}(w), \quad (5)$$

with another universal constant  $A_1 := R_\chi (R_\xi^+)^{d^*} / [(1 + \delta)R_c]$ , defined by other standard universal amplitudes [19]  $R_\xi^+ = (\alpha A_+)^{1/d^*} \xi_0^+$  and  $R_c = \alpha A_+ C_+ / B^2$ , where  $\alpha$  is the specific-heat exponent and  $A_+$  a nonuniversal specific-heat amplitude for  $t > 0$  and  $h = 0$ . Eqs. (4) and (5) determine completely universal  $W_{++}(y)$  within the local-functional approach.

Asymptotic behavior of  $W_{++}(y)$  as  $y \rightarrow \infty$  follows from Eqs. (4) and (5) by taking  $w \rightarrow 0$  from which Eq. (4) yields  $y^\nu \approx 2 \ln(B_+/w) + O(w)$ , where  $B_+$  is some universal constant. Similarly,  $W_{++}(y)$  as  $y \rightarrow -\infty$  is obtained from (4) and (5) by taking  $w \rightarrow 1$  yielding a similar expression for  $|y|^\nu$  but in terms of  $w - 1$ . Solving for  $w$  and substituting into Eq. (5) gives the following

$$W_{++}(y) \approx \begin{cases} -W_{+, \infty} y^{2-\alpha} \exp(-y^\nu), & \text{as } y \rightarrow +\infty; \\ -W_{-, \infty} |y|^{2-\alpha} \exp(-U_\xi |y|^\nu), & \text{as } y \rightarrow -\infty; \end{cases} \quad (6)$$

where  $U_\xi = \xi_0^+/\xi_0^-$ , and  $W_{\pm, \infty}$  are new universal amplitudes. The results summarized by Eq. (6) are *general* in that they hold in arbitrary spatial dimension  $d$ . Previous results, referring to some special cases, such as exact calculations on the Ising strip [7], and on the Ising chain subject to two identical surface fields, mean-field analysis based on the Ginzburg-Landau  $\varphi^4$  Hamiltonian [5], as well as mean-field treatments of confined fluids [8], confirm the power-law-exponential behavior of  $W_{++}(y)$  shown in Eq. (6).

In obtaining these results one can, to a very good approximation, set  $\mathcal{G}(x) = x^2$  [4]. This also applies to all subsequent results pertaining to the symmetric b.c. and greatly simplifies the calculations.

Mean-field form of  $W_{++}(y)$  in terms of the Jacobi functions [5] follows also within local-functional approach from Eqs. (4) and (5), when classical values for critical exponents are employed along with the scaling functions  $Y_{\pm}(\cdot)$  and  $Z_{\pm}(\cdot)$  for  $d \geq 4$ .

*Excess (Gibbs) adsorption*  $\Gamma(t, h)$ . The Gibbs adsorption, defined by  $\Gamma(t, h) = \int_0^L [m(z; t, h) - m_b(t, h)] dz$  is an integrated measure of the degree of ordering of spins or, equivalently, in the language of fluids, the amount of adsorbed substance on the walls [20]. From the above definition and the scaling postulate [3]  $m(z, L, T) \approx m_0(t) \psi_{++}(x, y)$ ,  $x := z/L$ , valid in the scaling limit

$t \rightarrow 0$ ,  $L \rightarrow \infty$ ,  $z \rightarrow \infty$ ,  $L - z \rightarrow \infty$ , follows:

$$\Gamma(t, 0) = B(\xi_0^+)^{\beta/\nu} L^{1-\beta/\nu} G(y), \quad (7a)$$

$$G(y) := |y|^\beta \int_0^1 [\psi_{++}(x, y) - \Theta(-y)] dx, \quad (7b)$$

with  $G(y)$  universal and  $\Theta(\cdot)$  the Heaviside function. Asymptotically,  $G(y) \sim |y|^{\beta-\nu}$  as  $|y| \rightarrow \infty$ , so that  $G(y)$  vanishes for large  $y$  and  $d < 4$ ,  $\beta < \nu$ . Since  $G(y)$  is not smooth at  $y = 0$ , we prefer to express results in terms of the universal quantity  $\tilde{G}(y)$ , defined for  $d < 4$  by

$$\tilde{G}(y) = G(y) + |y|^\beta \Theta(-y), \quad (8)$$

so that  $\int_0^L mdz = B(\xi_0^+)^{\beta/\nu} L^{1-\beta/\nu} \tilde{G}(y)$ . Local functional theory predicts that for  $d < 4$ :

$$\tilde{G}(y) = (1/A_2)y^{\beta-\nu} \int_w^\infty \frac{\sqrt{\frac{\tilde{Z}_\pm(u)}{\tilde{Y}_\pm(u)} du}}{u^{\nu/\beta} \sqrt{1 - (\frac{w}{u})^{1+\delta} \frac{\tilde{Y}_\pm(w)}{\tilde{Y}_\pm(u)}}} \quad (9)$$

For  $d \geq 4$  ( $\beta = \nu = 1/2$ ) scaling forms given by Eqs. (7a,7b) and (9) fail and one has to redefine them to encompass a logarithmic correction: in the limit  $L \rightarrow \infty$  and  $t \rightarrow 0$ ,  $\Gamma(L, T) \approx B\xi_0^+(K_1 \ln L + G(y)) + \Gamma_0(t)$ , with  $K_1$  as a universal constant, and  $\Gamma_0(t)$  a non-universal additive background containing both analytic background terms and singular corrections. Mean-field results, that follow from Eq. (9) when classical values of critical exponents are used along with the scaling forms  $\tilde{Y}_+(y) = 1 + 2/y^2$ ,  $\tilde{Y}_-(y) = (1 - 1/y^2)^2$ ,  $\tilde{Z}_\pm(u) = 1$  (one reads them off from Eqs. (3a, 3b) for  $d \geq 4$ ) can be expressed in terms of a elliptic integral of the first kind:

$$\tilde{G}(y) = \begin{cases} -\sqrt{2} \ln[2K^2(k)], & y = 4(2k^2 - 1)K^2(k), \\ & 1/2 \leq k^2 \leq 1; \\ -\sqrt{2} \ln[2(1 - k^2)K^2(k)], \\ y = -4(1 + k^2)K^2(k), & 0 \leq k \leq 1; \\ -\sqrt{2} \ln[2K^2(k)], & y = -4(1 - 2k^2)K^2(k), \\ & 0 \leq k^2 \leq 1/2. \end{cases} \quad (10)$$

Mean-field universal scaling function  $\tilde{G}(y)$  is shown by Fig. 2 below, together with the  $d = 3$  result.

To derive *quantitative* predictions at  $d = 3$  for  $W_{++}(y)$  and  $\tilde{G}(y)$  we need to substitute into Eqs. (4), (5) and (9) specific values for bulk critical exponents along with suitable choices for  $Y_\pm(y)$  and  $Z_\pm(y)$ . We represent bulk scaling functions using parametric models introduced by Schofield [21]. These have been developed further [18, 22] and are believed to give the best available fits to bulk data and, by their very construction, to give scaling functions satisfying required analyticity properties. For our purposes, pertaining to the present physical problem situated in a one-phase region, the original ‘‘linear’’ parametric model [12, 21] was found to suffice [23]. At  $d = 3$  we take  $\beta = 0.328$  and  $\nu = 0.632$  (all other exponents follow from the scaling relations) and a satisfactory fit

to the bulk amplitude ratios, being properties of bulk scaling functions, is provided by taking  $b^2 = 1.30$  and  $a_2 = 0.28$  in the notation of [22], in the linear model. The universal scaling function  $W_{++}(y)$  that follows from our calculations in  $d = 3$  is presented in Fig. 1 together with an earlier exact curve in spatial dimensions  $d = 2$  [7] and  $d \geq 4$  (mean-field) [5], which not being universal [17] is shown in reduced universal form  $W_{++}(y)/W_{++}(0)$ . The  $d = 3$  result confirms qualitatively similar structure

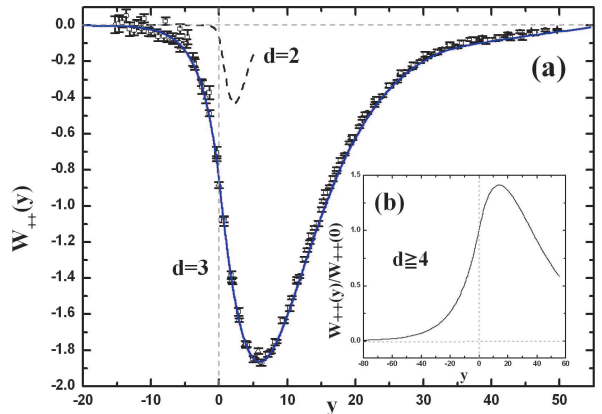


FIG. 1: Plots of the scaling function  $W_{++}(y)$  for the Ising universality class in (a)  $d = 3$  as obtained by local-functional theory (solid line) and compared with MC result [11] (open circles, rectangles and triangles); exact  $d = 2$  (dashed line) [7]; (b)  $d \geq 4$ , presented in a reduced (universal) form derived for the first time in [5].

of the CF in  $d = 3$  with the ones observed in all the other spatial dimensions. This refers to the negative sign of the CF for like b.c., smoothness across the whole interval of scaling variable  $y \in \mathbb{R}$  (apart from an  $|y|^{2-\alpha}$  singularity at  $y = 0$ , which can be shown to be quite general) as expected based on the fact that the critical point of the film ( $T_c(L)$ ,  $h_c(L)$ ) is located *off* the temperature axis at a non-zero critical bulk field [24]. It also follows from this analysis that the minimum of  $W_{++}(y)$  in  $d = 3$  is located *above* the critical point as in other dimensions. Fig. 1 shows that there is striking agreement between the present calculation of  $W_{++}(y)$  and recent MC simulation results [11], with quoted value of  $W_{++}^{\text{MC}}(0) = -0.884$  implying that the value of the Casimir amplitude  $A_{++}^{\text{MC}} = -0.442$  is closer to this and the earlier local-functional result of  $-0.42(8)$  [4] as compared to the previous MC study [5].

Numerical predictions for  $\tilde{G}(y)$  in  $d = 3$ , based on the EdGF Eqs. (4) and (9) within a parametric representation, as well as analytic result in the mean-field limit according to Eq. (10), are given by Fig. 2, showing smooth curves, diverging as  $|y|^\beta$  for  $y \rightarrow -\infty$  in accord with the general definition of  $\tilde{G}(y)$ .

There is also much experimental and theoretical interest in the Casimir effect for the antisymmetric b.c. (+-) with recent MC results presented for  $W_{+-}(y)$  [11]. In this case, complications arise in the application of local-

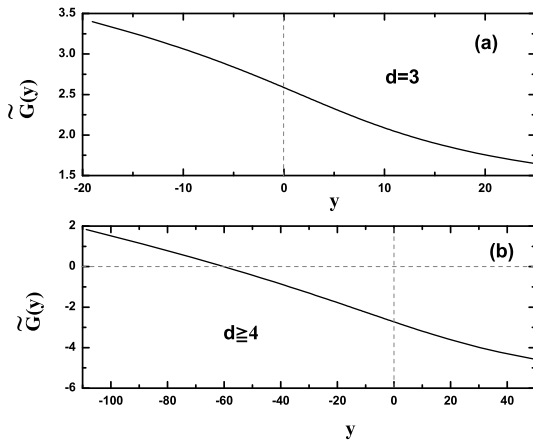


FIG. 2: Universal scaling function  $\tilde{G}(y)$  of the excess adsorption  $\Gamma$  for the Ising universality class calculated within local-functional theory (a) in  $d=3$ , by Eqs. (4) and (9); (b) in  $d \geq 4$  according to the EdGF analytic solution (10).

functional methods for two main reasons: (i) the approximation  $\mathcal{G}(x) = x^2$  no longer holds and one needs to use the far more complicated form of  $\mathcal{G}(x)$  as introduced in [4]; (ii) one needs to extend the bulk scaling functions  $Y_-(\cdot)$ ,  $Z_-(\cdot)$  into the two-phase region, a somewhat ad hoc procedure although possible if one uses trigonometric parametric models (instead of the linear model) [18, 22] giving rise to “nonclassical van der Waals loops”. However, this more complicated calculation is possible and forms the subject of ongoing research.

More details will follow in a longer report. We kindly thank Prof.S. Dietrich and Dr.O. Vasilyev for making their MC data [11] available for us that enabled comparisons with the present result of EdGF theory within Fig. 1.

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