

# Nonuniversal size dependence of the free energy of confined systems near criticality

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(Received 10 August 2001; published 2 July 2002)

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 leading nonuniversal  $L$  dependence of  $f_s$  near  $T_c$ . This implies a significant restriction for the validity of universal finite-size scaling for model systems and real systems. For film geometry we predict a leading nonuniversal contribution to the critical Casimir force above the superfluid transition of  $^4\text{He}$ .

DOI: 10.1103/PhysRevE.66.016102

PACS number(s): 05.70.Jk, 64.60.-i

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 [1–3]. 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  $f_b \equiv f(t, \infty)$  can be decomposed as

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

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 [4,5], the singular part of the finite-size free-energy density may be defined by

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

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 asymptotic structure [4–6]

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

where  $\mathcal{F}(x)$  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  $\tilde{a}$  of lattice models, the inverse cutoff  $\Lambda^{-1}$  of field theories, or the length scale of subleading long-range interactions). Equation (3) includes the bulk limit  $f_s(t, \infty) = f_{bs}(t) = Y\xi^{-d}$  with a universal amplitude  $Y$ . Equations (1)–(3) are also expected to remain valid for nonperiodic boundary conditions and noncubic geometries provided that  $f_0(t)$  in Eq. (2) is replaced by the nonsingular part  $f_{ns}(t, L)$  with a regular  $t$  dependence [5], where now the scaling function  $\mathcal{F}(x)$  depends on the boundary conditions, the geometry, and on the universality class of the bulk critical point, but not on  $\tilde{a}$  or  $\Lambda$  and not on other interaction details [4–6]. In particular, subleading long-range interactions (such as van der Waals forces in fluids) that do not

affect the universal bulk critical behavior of  $f_{bs}(t)$  are assumed to contribute only to the regular part  $f_{ns}(0, L)$  or  $f_{ns}(t, L)$ , but not to the singular part  $f_s(t, L)$  [7–10].

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  $F$  in film geometry [7–11]

$$F = -\partial f^{ex}(t, L)/\partial L, \quad (4)$$

where the excess free energy per unit area is given by

$$f^{ex}(t, L) = Lf(t, L) - Lf_b(t). \quad (5)$$

Equations (1)–(5) yield the singular part of  $F = F_s + F_{ns}$ ,

$$F_s(\xi, L) = L^{-d} X(L/\xi), \quad (6)$$

where

$$X(x) = (d-1)\mathcal{F}(x) - x\mathcal{F}'(x) + Yx^d, \quad (7)$$

with  $\mathcal{F}'(x) = \partial\mathcal{F}(x)/\partial x$ . The universal scaling structure of Eqs. (3), (6), and (7) has been confirmed by renormalization-group (RG) and model calculations [7,12,13]. In particular, quantitative predictions of  $X(x)$  for Dirichlet boundary conditions (DBC) [7] that are relevant for the superfluid transition of  $^4\text{He}$  [14] have been used in the analysis of experimental data [9,15].

In this paper we show that the conditions for the validity of finite-size universality of  $f_s$  and  $F_s$  are significantly more restricted than those for bulk universality of  $f_{bs}$ . On the basis of exact and approximate results within the  $\varphi^4$  field theory we shall analyze the effect of two sources in the  $\varphi^4$  Hamiltonian that have recently been shown [16–18] to cause nonscaling finite-size effects on the susceptibility  $\chi$  for PBC: (i) a short-range interaction term  $\sim \mathbf{k}^2$  with a sharp cutoff  $\Lambda$  in  $\mathbf{k}$  space, and (ii) an additional subleading long-range interaction term  $\sim b|\mathbf{k}|^\sigma$ ,  $2 < \sigma < 4$ .

The size dependence of  $f$  near  $T_c$  is more complex than that of  $\chi$  [5]. *A priori* it is not obvious whether or not and to what extent nonuniversal effects enter the singular (rather than regular) part of  $f(t, L)$ . In particular, this is an open

question for case (ii) with *nonperiodic* boundary conditions. Here we shall consider both PBC and DBC and shall show that the singular parts  $f_s$  and  $F_s$  are significantly affected by nonuniversal nonscaling terms.

Specifically, we find for  $2 < d < 4$  that Eqs. (3) and (6) must be complemented as

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

for case (i), and

$$F_s(\xi, L, b) = -bL^{-d+2-\sigma} B(L/\xi) + L^{-d} X(L/\xi) \quad (9)$$

for case (ii), respectively, where the function  $\Phi$  has a finite critical value  $\Phi(0) > 0$  and where the function  $B(L/\xi)$  has a *nonexponential* decay  $\sim (L/\xi)^{-2}$  above  $T_c$  for both PBC and DBC. This implies (i) that the nonscaling  $L^{-2}$  term in Eq. (8) exhibits a dominant size dependence compared to the  $L^{-d}$  scaling term and (ii) that the nonuniversal term proportional to  $b$  in Eq. (9) implies an algebraic  $L$  dependence  $\sim b\xi^2 L^{-d-\sigma}$  that dominates the *exponential* finite-size scaling term of  $X$  above  $T_c$  for both PBC and DBC. By contrast, for the  $\varphi^4$  lattice model with short-range interaction and PBC, we find that Eqs. (3) and (6) are indeed valid except that for  $L \gg \xi$  above  $T_c$  the exponential scaling arguments of  $\mathcal{F}$  and  $X$  must be formulated in terms of the lattice-dependent ‘‘exponential’’ correlation length [19,20].

The new nonscaling finite-size effect (i) exhibited in Eq. (8) is pertinent to the entire  $\xi^{-1} - L^{-1}$  plane. In particular, it exists at  $T_c$ , where it implies the nonuniversality of the critical Casimir force  $L^{-2} \Lambda^{d-2} \Phi(0)$  in film geometry. Furthermore, the new nonscaling effect (ii) exhibited in Eq. (9) 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 [16–18]. The latter are of limited physical relevance since in real systems they are dominated by the surface terms of  $O(L^{-1})$ . In this paper we predict a leading nonuniversal nonscaling effect on the singular part  $F_s$  (rather than on the regular part  $F_{ns}$  [7]), not only for model systems with PBC but also for real systems with DBC.

We start from the standard  $\varphi^4$  continuum Hamiltonian

$$H = \int d^d x \left[ \frac{1}{2} r_0 \varphi^2 + \frac{1}{2} (\nabla \varphi)^2 + u_0 (\varphi^2)^2 \right] \quad (10)$$

with  $r_0 = r_{0c} + a_0 t$  for the  $n$ -component field  $\varphi(\mathbf{x})$  in a partially confined  $L^{d'} \times \infty^{d-d'}$  geometry. This model requires a specification of the  $\mathbf{x}$  dependence of  $\varphi(\mathbf{x})$  at short distances. We decompose the vector  $\mathbf{x}$  as  $(\mathbf{y}, \mathbf{z})$ , where  $\mathbf{z}$  denotes the coordinates in the  $d' < d$  confined directions. We consider two cases.

*Case (i).* We assume PBC and a sharp cutoff  $\Lambda$ , i.e., we assume that the Fourier amplitudes  $\hat{\varphi}_{\mathbf{p}, \mathbf{q}}$  of  $\varphi(\mathbf{y}, \mathbf{z}) = L^{-d'} \sum_{\mathbf{p}} \int_{\mathbf{q}} \hat{\varphi}_{\mathbf{p}, \mathbf{q}} e^{i(\mathbf{p} \cdot \mathbf{z} + \mathbf{q} \cdot \mathbf{y})}$  are restricted to wave vectors  $\mathbf{p}$  and  $\mathbf{q}$  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  $(2\pi)^{-d+d'} \int d^{d-d'} q$ , and  $\sum_{\mathbf{p}}$  runs over  $p_j = 2\pi m_j / L$  with  $m_j = 0, \pm 1, \pm 2, \dots$

The question can be raised whether or not there exists a non-negligible cutoff dependence of the finite-size free-energy density per component (divided by  $k_B T$ ),

$$f_{d,d'}(t, L, \Lambda) = -n^{-1} L^{-d'} \lim_{\tilde{L} \rightarrow \infty} \tilde{L}^{-d+d'} \ln Z_{d,d'}, \quad (11)$$

where

$$Z_{d,d'}(t, L, \tilde{L}, \Lambda) = \prod_{\mathbf{k}, \mathbf{q}} \int \frac{d\hat{\varphi}_{\mathbf{p}, \mathbf{q}}}{\Lambda^{(d-2)/2}} \exp(-H) \quad (12)$$

is the dimensionless partition function of a  $L^{d'} \times \tilde{L}^{d-d'}$  geometry. For comparison we shall also consider the free-energy density  $\hat{f}(t, L, \tilde{a})$  of the  $\varphi^4$  lattice model

$$\hat{H} = \tilde{a}^d \left[ \sum_i \left( \frac{r_0}{2} \varphi_i^2 + u_0 (\varphi_i^2)^2 \right) + \sum_{\langle ij \rangle} \frac{J}{2\tilde{a}^2} (\varphi_i - \varphi_j)^2 \right] \quad (13)$$

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  $\hat{H}$ .

We shall answer this question in the exactly solvable limit  $n \rightarrow \infty$  at fixed  $u_0 n$  where the free-energy density is [21]

$$f_{d,d'}(t, L, \Lambda) = -\frac{1}{2} \Lambda^d \ln \pi - \frac{(r_0 - \chi^{-1})^2}{16u_0 n} + \frac{1}{2} L^{-d'} \sum_{\mathbf{p}} \int_{\mathbf{q}} \ln [\Lambda^{-2} (\chi^{-1} + \mathbf{p}^2 + \mathbf{q}^2)]. \quad (14)$$

Here  $\chi^{-1}$  is determined implicitly by

$$\chi^{-1} = r_0 + 4u_0 n L^{-d'} \sum_{\mathbf{p}} \int_{\mathbf{q}} (\chi^{-1} + \mathbf{p}^2 + \mathbf{q}^2)^{-1}. \quad (15)$$

The bulk free energy  $f_b$  and bulk susceptibility  $\chi_b$  above  $T_c$  are obtained by the replacement  $L^{-d'} \sum_{\mathbf{p}} \int_{\mathbf{q}} \rightarrow \int_{\mathbf{k}}$ , and the critical point is determined by  $r_0 = r_{0c} = -4u_0 n \int_{\mathbf{k}} \mathbf{k}^{-2}$  where  $\mathbf{k} \equiv (\mathbf{p}, \mathbf{q})$ . The bulk correlation length above  $T_c$  is  $\xi = \chi_b^{1/2} = \xi_0 t^{-\nu}$  where  $\nu = (d-2)^{-1}$ . The regular part of  $f_b$  reads  $f_0 = \tilde{c}_1 \Lambda^d - r_0^2 / (16u_0 n)$  where  $\tilde{c}_1$  is a  $d$  dependent constant. The singular part of  $f_b$  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 nonscaling part

$$\Phi_{d,d'}(\xi^{-1} \Lambda^{-1}) = \frac{d'}{6(2\pi)^{d-2}} \int_0^\infty dy \left[ \int_{-1}^1 dq e^{-q^2 y} \right]^{d-1} \times \exp[-(1 + \xi^{-2} \Lambda^{-2}) y] \quad (16)$$

and the subleading universal scaling part

$$\begin{aligned} \mathcal{F}_{d,d'}(L/\xi) &= \frac{A_d}{2(4-d)} \left[ (L/\xi)^{d-2} P^2 - \frac{2}{d} P^d \right] \\ &+ \frac{1}{2} \int_0^\infty \frac{dy}{y} \left( \sqrt{\frac{\pi}{y}} \right)^{d-d'} W_{d'}(y) e^{-P^2 y/4\pi^2}, \end{aligned} \quad (17)$$

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

$$\begin{aligned} P^{d-2} &= (L/\xi)^{d-2} - \frac{4-d}{4\pi^2 A_d} \int_0^\infty dy \left( \sqrt{\frac{\pi}{y}} \right)^{d-d'} \\ &\times W_{d'}(y) e^{-P^2 y/4\pi^2}, \end{aligned} \quad (18)$$

$$W_d(y) = \left( \sqrt{\frac{\pi}{y}} \right)^d - \left( \sum_{m=-\infty}^{\infty} e^{-ym^2} \right)^d. \quad (19)$$

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  $-\xi^{-2}\Lambda^{-2}y$  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  $\Phi_{d,d'}(\xi^{-1}\Lambda^{-1})$  as in Eq. (16). This proves that the *singular* part of  $f(t,L)$  has a nonuniversal nonscaling form for the  $\varphi^4$  field theory with PBC and with a sharp cutoff. A detailed derivation of Eqs. (16)–(19) will be given elsewhere [22].

These results have a significant consequence for the critical Casimir effect. Instead of Eq. (6) we obtain from Eqs. (8) and (16)–(19) in film geometry ( $d' = 1$ )

$$F_s(\xi, L, \Lambda) = L^{-2}\Lambda^{-2}\Phi_{d,1}(\xi^{-1}\Lambda^{-1}) + L^{-d}X(L/\xi). \quad (20)$$

Thus, the *singular* part of the critical Casimir force has a *leading* nonuniversal term  $\sim L^{-2}$ , in addition to the *subleading* universal terms  $\sim L^{-d}$  of previous theories [7–10,12], both for  $T \geq T_c$  and for  $T < T_c$ .

We have also calculated  $f_{d,d'}$  and  $F_s$  for the lattice Hamiltonian (13) and for the continuum Hamiltonian (10) with a *smooth* cutoff in the large- $n$  limit. In both cases the scaling form (3) is found to be valid. For the lattice model, however, the second-moment bulk correlation length  $\xi$  in the argument of  $\mathcal{F}$  must be replaced by the lattice-dependent exponential correlation length [19,20]. Specifically, we find, at fixed  $t > 0$ , the exponential large- $L$  behavior

$$\hat{f}_s(t, L, \tilde{a}) - f_{bs} = -d'(L/2\pi\xi_1)^{(d-1)/2} L^{-d} \exp(-L/\xi_1), \quad (21)$$

where  $\xi_1 = (\tilde{a}/2)[\operatorname{arcsinh}(\tilde{a}/2\xi)]^{-1}$  is the exponential correlation length in the direction of one of the cubic axes. Note that the *nonuniversal* dependence of  $\xi_1$  on  $\tilde{a}$  is nonnegligible in the exponent of Eq. (21) [19].

The sensitivity of  $f_s(t, L, \Lambda)$  and  $F_s$  with respect to the cutoff procedure can be explained in terms of a corresponding sensitivity of the *bulk* correlation function  $G(\mathbf{x}) = \langle \varphi(\mathbf{x})\varphi(0) \rangle$  in the range  $|\mathbf{x}| \gg \xi$  [19]. For example, for the

$\varphi^4$  continuum Hamiltonian (10) with an isotropic sharp cutoff  $|\mathbf{k}| \leq \Lambda$  we find, in the large- $n$  limit, the oscillatory power-law decay above  $T_c$ ,

$$\begin{aligned} G(\mathbf{x}) &= 2\Lambda^{d-2} (2\pi x \Lambda)^{-(d+1)/2} \frac{\sin[\Lambda x - \pi(d-1)/4]}{1 + \xi^{-2}\Lambda^{-2}} \\ &+ O(e^{-x/\xi}), \end{aligned} \quad (22)$$

for large  $x = |\mathbf{x}| \gg \xi$  corresponding to the existence of long-range spatial correlations that dominate the exponential scaling dependence  $\sim e^{-x/\xi}$ . By contrast,  $G(\mathbf{x})$  has an exponential decay for the lattice model (13) with purely short-range interaction [19]. An exponential decay of  $G(\mathbf{x})$  is also valid for the continuum model (10) with a smooth cutoff [19].

The nonuniversal cutoff effects on  $f_s$ ,  $F_s$ , and  $G(\mathbf{x})$  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 also as a signal for a restriction of finite-size universality in physical systems. We substantiate this interpretation by demonstrating that a corresponding reduction of the finite-size scaling regime 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  $b|\mathbf{k}|^\sigma$  with  $2 < \sigma < 4$ , in addition to the short-range term  $\mathbf{k}^2$ . It is well known that the subleading interaction  $\sim |\mathbf{k}|^\sigma$  corresponds to a spatial interaction potential  $V(\mathbf{x}) \sim |\mathbf{x}|^{-d-\sigma}$  that does not change the universal bulk critical behavior [23]. Interactions of this type exist in real fluids. As pointed out by Dantchev and Rudnick [18], the presence of this interaction yields leading nonscaling 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 [16,17].

In real systems with nonperiodic boundary conditions, however, these nonscaling finite-size effects become only subleading corrections 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  $F_s$ , 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  $\varphi(\mathbf{y}, 0) = \varphi(\mathbf{y}, L) = 0$ , i.e., we assume that  $\Sigma_p$  in the Fourier representation of  $\varphi(\mathbf{y}, z) = L^{-1} \sum_p \int_{\mathbf{q}} \hat{\varphi}_{p,\mathbf{q}} e^{i\mathbf{q}\cdot\mathbf{y}} \sin(pz)$  runs over  $p = \pi m/L$ ,  $m = 1, 2, \dots$ . The presence of the subleading interaction  $b|\mathbf{k}|^\sigma$  implies, for  $L \gg \xi$  above  $T_c$ , a nonuniversal term  $\sim b$  in

$$f_s(t, L, b) - \tilde{f} = -bL^{-d+2-\sigma} \Psi(L/\xi) + L^{-d} \mathcal{G}(L/\xi), \quad (23)$$

where  $\mathcal{G}(L/\xi)$  is the known universal scaling function for purely short-range interaction with an *exponential* large- $L$  behavior [7]. By contrast we find that  $\Psi(L/\xi)$  has an algebraic  $L$  dependence. Performing a one-loop RG calculation we obtain

$$\Psi(L/\xi) = \frac{1}{2} (2\pi)^{\sigma-4} \int_{(L/\xi)^2}^{\infty} dx \left( 1 + x \frac{\partial}{\partial x} \right) \tilde{\Psi}(x), \quad (24)$$

$$\begin{aligned} \tilde{\Psi}(x) = & \int_0^{\infty} dy y^{(2-\sigma)/2} e^{-xy/4\pi^2} \left( \sqrt{\frac{\pi}{y}} \right)^{d-1} \\ & \times \tilde{W}_1(y) \gamma^* \left( \frac{2-\sigma}{2}, -\frac{xy}{4\pi^2} \right), \end{aligned} \quad (25)$$

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

$$\tilde{W}_1(y) = \sqrt{\frac{\pi}{y}} - \frac{1}{2} \sum_{n=-\infty}^{\infty} \exp\left(-\frac{y}{4} n^2\right). \quad (26)$$

We have found that cutoff effects are negligible for the function  $\Psi(L/\xi)$ . At fixed  $\xi$ , the large- $L$  behavior is  $\Psi(L/\xi) \sim (L/\xi)^{-2}$ . Equation (23) yields the following form:

$$B(L/\xi) = (d-3+\sigma)\Psi(L/\xi) - (L/\xi)\Psi'(L/\xi) \quad (27)$$

for the nonuniversal contribution to  $F_s$  in Eq. (9). The crucial

consequence is that the leading critical temperature dependence  $\sim b\xi^2 L^{-d-\sigma}$  of  $F_s$  for  $L \gtrsim \xi$  above  $T_c$  is algebraic and nonuniversal, whereas the critical temperature dependence of the scaling part  $X(L/\xi)$  derived from  $\mathcal{G}(L/\xi)$  is exponential and universal [7]. This prediction is applicable to  ${}^4\text{He}$  above  $T_\lambda$  after specification of the interaction parameters  $b$  and  $\sigma$ , and may have significant consequences for the interpretation of existing [15] and future experimental data. A detailed derivation of Eqs. (23)–(27) will be given elsewhere [22].

Finally, for comparison we present our result for  $\Psi(L/\xi)$  for film geometry in the presence of PBC. In one-loop order we obtain for  $\Psi_{PBC}(L/\xi)$  the same form as given for  $\Psi(L/\xi)$  in Eqs. (24) and (25) but with  $\tilde{W}_1(y)$  replaced by  $W_1(y)$ , Eq. (19). For the large- $L$  behavior we find  $\Psi_{PBC}(L/\xi) \sim (L/\xi)^{-2}$ , which dominates the exponential scaling dependence of  $X$ . This is parallel to the algebraic decay of  $G(\mathbf{x})$  in the presence of van der Waals type interactions [24]. Our prediction of a nonexponential nonscaling effect on  $F_s$  above  $T_c$  for PBC can be tested by Monte Carlo simulations [9].

Support by DLR and by NASA under Contract Nos. 50WM9911 and 1226553 is acknowledged.

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