

Nature of Asymmetry in Fluid Criticality

Mikhail A. Anisimov* and Jingtao Wang

Department of Chemical and Biomolecular Engineering and Institute for Physical Science and Technology, University of Maryland, College Park, Maryland 20742, USA

(Received 28 April 2006; published 14 July 2006)

By combining accurate liquid-vapor coexistence and heat-capacity data, we have unambiguously separated two nonanalytical contributions of liquid-gas asymmetry in fluid criticality and showed the validity of “complete scaling” [Fisher *et al.*, Phys. Rev. Lett. **85**, 696 (2000); Phys. Rev. E **67**, 061506 (2003)]. We have also developed a method to obtain two scaling-field coefficients, responsible for the two sources of the asymmetry, from mean-field equations of state. Since the asymmetry effects are completely determined by Ising critical exponents, there is no practical need for a special renormalization-group theoretical treatment of asymmetric fluid criticality.

DOI: 10.1103/PhysRevLett.97.025703

PACS numbers: 64.60.Fr

A controversial issue of liquid-gas asymmetry in fluids has been a subject of prolonged discussions for more than a century since Cailletet and Mathias’s discovery of the empirical “law” of rectilinear diameter [1]. According to this law, the mean of the densities of liquid ρ' and saturated vapor ρ'' is a linear function of the temperature T :

$$\Delta\hat{\rho}_d \equiv \frac{\rho' + \rho''}{2\rho_c} = 1 + D|\Delta\hat{T}|, \quad (1)$$

where ρ_c is the critical density; $\Delta\hat{T} = (T - T_c)/T_c$ is the reduced distance to the critical temperature T_c . The system-dependent coefficient D generally increases from 0.02 for ^3He [2] to values larger than unity with increase of T_c [3,4]. In this Letter, we demonstrate how the mean-field rectilinear diameter splits up in the critical region into two “singular diameters” associated with two different sources of asymmetry. It is commonly accepted that fluids asymptotically ($\Delta\hat{T} \rightarrow 0$) belong to the critical-point universality class of the Ising model [5]. We argue that the asymmetry effects in near-critical fluids, at least in the lower approximation, are determined by Ising critical exponents; hence, in contrast to a commonly used approach, there may be no need for a special renormalization-group theoretical treatment of asymmetric fluid criticality.

Thermodynamics near a critical point is controlled by two scaling fields, “ordering” h_1 and “thermal” h_2 , while an appropriate field-dependent potential h_3 is a universal function of h_1 and h_2 [5]:

$$h_3 \simeq h_2^{2-\alpha} f^\pm\left(\frac{h_1}{h_2^{\beta+\gamma}}\right), \quad (2)$$

where $\alpha = 0.109$, $\beta = 0.326$, and $\gamma = 1.239$, interrelated as $\alpha + 2\beta + \gamma = 2$, are universal Ising critical exponents in the scaling power laws (as a function of h_2 at $h_1 = 0$) for the weakly divergent susceptibility, order parameter, and strongly divergent susceptibility, respectively [5]. The scaling “densities,” a strongly fluctuating order parameter ϕ_1 and a weakly fluctuating ϕ_2 , conjugate to h_1 and h_2 , such

that $dh_3 = \phi_1 dh_1 + \phi_2 dh_2$. The universal function f^\pm contains two system-dependent amplitudes, and the superscript \pm refers to $h_2 \gtrless 0$. The Ising model formulated for fluids is known as “lattice gas” [6]. In the lattice gas, h_3 is the “critical part” of the grand thermodynamic potential $\Omega = -PV$, taken per unit volume; thus, h_1 is the dimensionless chemical-potential difference $\Delta\hat{\mu} = (\mu - \mu_c)/k_B T_c$, where μ_c is the value of the chemical potential at the critical point and k_B is Boltzmann’s constant. By definition, $h_1 = 0$ along the critical isochore above T_c and along the liquid-vapor coexistence curve below T_c . Hence, in the lattice gas the order parameter $\phi_1 = \Delta\hat{\rho} = (\rho - \rho_c)/\rho_c$, the thermal scaling field $h_2 = \Delta\hat{T}$, and $\phi_2 = \Delta(\hat{\rho}\hat{S}) = (\hat{\rho}\hat{S} - \hat{\rho}_c\hat{S}_c)/k_B$, where $\hat{\rho}\hat{S}$ is the density of entropy.

The lattice gas has a special symmetry: The order parameter is symmetric with respect to the critical isochore ($D = 0$). Since the early 1970s, the liquid-gas asymmetry has been commonly incorporated into the lattice-gas analogy by linear mixing of two independent physical fields $\Delta\hat{\mu}$ and $\Delta\hat{T}$ into both the theoretical scaling fields h_1 and h_2 [7]. Since the absolute value of entropy is arbitrary, mixing of $\Delta\hat{T}$ into h_1 plays no role. Contrarily, mixing of $\Delta\hat{\mu}$ (with b_2 as a mixing coefficient) into h_2 has an important consequence, known as the “singular diameter”: the mean of the densities must contain a nonanalytic contribution $\propto b_2|\Delta\hat{T}|^{1-\alpha}$, so that $d(\Delta\hat{\rho}_d)/d\hat{T} \propto b_2|\Delta\hat{T}|^{-\alpha}$, diverging weakly. However, the chemical potential would remain an analytical function of temperature along the liquid-vapor coexistence and the symmetry would be restored by a redefinition of the order parameter as $\phi_1 = \Delta\hat{\rho} + b_2\Delta(\hat{\rho}\hat{S})$.

At this point, we encounter a major problem. First of all, the existence of the $|\Delta\hat{T}|^{1-\alpha}$ term in the “diameter” of real fluids has never been detected unambiguously. While some fluids show strong deviations from rectilinear diameter, apparently even stronger than $|\Delta\hat{T}|^{1-\alpha}$ [3], many fluids show very little or no deviation at all [8,9]. Moreover, there

is a conceptual problem with mapping real fluids into the lattice gas even at the mean-field level. In the mean-field approximation, the critical part of the thermodynamic potential is represented by a Landau expansion:

$$h_3 = \frac{1}{2}a_0 h_2 \phi_1^2 + \frac{1}{4}u_0 \phi_1^4 - h_1 \phi_1. \quad (3)$$

When $h_1 = \Delta\hat{\mu}$, $h_2 = \Delta\hat{T} + b_2\Delta\hat{\mu}$, and $\phi_1 = \Delta\hat{\rho} + b_2\Delta(\hat{\rho}\hat{S})$, this expansion generates asymmetric terms $\propto b_2\Delta\hat{T}(\Delta\hat{\rho})^3$ and $\propto b_2(\Delta\hat{\rho})^5$. However, in the simplest equation of state that describes real-fluid behavior, the van der Waals equation, the term $\propto \Delta\hat{T}(\Delta\hat{\rho})^3$ is absent, while the term $\propto (\Delta\hat{\rho})^5$ exists. Furthermore, in most classical equations of state $d\hat{\mu}^2/d\hat{T}^2$ along the liquid-vapor coexistence exhibits a discontinuity directly related to the existence of the independent 5th-order term [10] in Landau expansion. The existence of the independent 5th-order term makes exact mapping of fluids into the lattice-gas model by the conventional mixing of physical fields impossible. On the other hand, a renormalization-group treatment of the 5th-order term resulted in an independent critical exponent $\theta_5 \approx 1.3$ [11] (not existing in the Ising model). We show, however, that asymmetric fluids can be consistently mapped into Ising criticality by applying so-called ‘‘complete scaling’’ originally proposed by Fisher and Orkoulas [12]. A redefinition of the order parameter, suggested by complete scaling, results in elimination of the 5th-order term in Landau expansion, thus making the renormalization-group treatment of the 5th-order term irrelevant for fluids.

Complete scaling suggests that all three physical fields $\Delta\hat{\mu}$, $\Delta\hat{T}$, and $\Delta\hat{P} = (P - P_c)/\rho_c k_B T_c$ are equally mixed into three scaling fields h_1 , h_2 , and h_3 . In the linear approximation

$$h_1 = a_1\Delta\hat{\mu} + a_2\Delta\hat{T} + a_3\Delta\hat{P}, \quad (4)$$

$$h_2 = b_1\Delta\hat{T} + b_2\Delta\hat{\mu} + b_3\Delta\hat{P}, \quad (5)$$

$$h_3 = c_1\Delta\hat{P} + c_2\Delta\hat{\mu} + c_3\Delta\hat{T}. \quad (6)$$

Before we apply complete scaling to describe asymmetry in fluids, we note that the relations between scaling and physical fields can be simplified. The coefficients a_1 and b_1 can be absorbed in two amplitudes in the scaling function f^\pm . The coefficients c_1 and c_2 are absorbed by making the thermodynamic potential h_3 dimensionless. The coefficient $c_3 = \hat{S}_c$ is determined by the choice of the critical value

of entropy. By adopting $\hat{S}_c = (k_B \rho_c)^{-1}(\partial P/\partial T)_{h_1=0,c} = (d\hat{P}/d\hat{T})_{\text{exc,c}}$, the slope of the saturation-pressure curve at the critical point, one obtains $a_2 = -a_3(d\hat{P}/d\hat{T})_{\text{exc,c}}$ and $b_3 = 0$. Finally, the scaling fields contain only two amplitudes responsible for asymmetry in fluid criticality:

$$h_1 = \Delta\hat{\mu} + a_3[\Delta\hat{P} - (d\hat{P}/d\hat{T})_{\text{exc,c}}\Delta\hat{T}], \quad (7)$$

$$h_2 = \Delta\hat{T} + b_2\Delta\hat{\mu}, \quad (8)$$

$$h_3 = -\Delta\hat{P} + \Delta\hat{\mu} + (d\hat{P}/d\hat{T})_{\text{exc,c}}\Delta\hat{T}. \quad (9)$$

As a result, while the order parameter in fluids is, in general, a nonlinear combination of density and entropy $\phi_1 = [\Delta\hat{\rho} + b_2\Delta(\hat{\rho}\hat{S})]/(1 + a_3\Delta\hat{\rho})$, the weakly fluctuating scaling density ϕ_2 in the first approximation is associated with the density of entropy only, $\phi_2 = \Delta(\hat{\rho}\hat{S})$. There are two important thermodynamic consequences of complete scaling that can be checked experimentally. First, the diameter ρ_d should contain two nonanalytical contributions, associated with the terms $a_3\Delta\hat{P}$ and $b_2\Delta\hat{\mu}$ in the scaling fields:

$$\begin{aligned} \hat{\rho}_d - 1 &= a_3(1 + a_3)\phi_1^2 + b_2\phi_2 + \dots \\ &= D_1|\Delta\hat{T}|^{2\beta} + D_2|\Delta\hat{T}|^{1-\alpha} + D_3|\Delta\hat{T}| + \dots, \end{aligned} \quad (10)$$

where $D_1 = a_3 B_0^2/(1 + a_3)$ and $D_2 = b_2 A_0^-/(1 - \alpha)$, with B_0 and A_0^- being the amplitudes in the asymptotic scaling power laws for the liquid or vapor densities, $\Delta\hat{\rho} = \pm B_0|\Delta\hat{T}|^\beta + \dots$, and isochoric heat capacity in the two-phase region, $C_V/k_B = A_0^-|\Delta\hat{T}|^{-\alpha} + \dots$ ($A_0^- = A_0^+/0.523$) [5(c)]. Note that, since $2\beta < 1 - \alpha$, the term $D_1|\Delta\hat{T}|^{2\beta}$ should dominate near the critical point. Second, the presence of this term implies a so-called Yang-Yang anomaly: The divergence of the heat capacity in the two-phase region is shared among the second derivatives of pressure and chemical potential [12]. Experimental verification of complete scaling is a very challenging task. The nonanalytical contributions in the diameter are usually not large enough to be separated unambiguously. Attempts to fit some experimental and simulation data to Eq. (10) showed very poor conversions [13], mainly because of a strong correlation between the linear and $D_2|\Delta\hat{T}|^{1-\alpha}$ terms. Experimental tests of the Yang-Yang anomaly are even more controversial since traces of impurities can easily mimic such an anomaly, thus making any conclusions unreliable [14].

We have been able to reliably determine the two asymmetry coefficients a_3 and b_2 and to conclusively prove the validity of complete scaling by combining accurate experimental and simulation liquid-vapor coexistence and heat-capacity data. We have exploited the fact that the coefficients D_2 and D_3 in Eq. (10) are actually coupled. As the weakly fluctuating scaling density ϕ_2 is the critical part of the entropy density, in the two-phase region at average density $\rho = \rho_c$

$$\phi_2 = \int \frac{C_V^{\text{cr}}}{k_B T} dT = -\frac{A_0^-}{1 - \alpha}|\Delta\hat{T}|^{1-\alpha} + B_{\text{cr}}|\Delta\hat{T}|, \quad (11)$$

where C_V^{cr} is the critical part of the isochoric heat capacity and B_{cr} the so-called ‘‘critical background,’’ fluctuation-induced analytical part of the heat capacity. The critical

background can be obtained from a ratio between B_{cr} , A_0^+ , and the nonasymptotic heat-capacity amplitude A_1^+ [15] and by subtracting the “ideal gas” from the total heat-capacity background. Both procedures yield very similar values of B_{cr} . Since $D_3 = -b_2 B_{cr}$, Eq. (10) contains only two adjustable coefficients, a_3 and b_2 . We have examined a number of systems, real fluids and simulated models [3,16], for which we could find both heat-capacity and coexistence data in the range $|\Delta\hat{T}| < 0.01$. In this range, the terms of higher order than linear in Eq. (10) are within experimental errors. Experimental data closer than $|\Delta\hat{T}| < 10^{-4}$ were avoided as they might be affected by errors in ρ_c and T_c and by other factors, such as gravity, impurities, etc. For all systems studied, we have been able to obtain reliable values of B_{cr} and conclusively separate two singular contributions to the diameter. Two typical examples that represent two different kinds of asymmetry in fluid criticality are shown in Fig. 1.

In diameters of some fluids, such as SF_6 , $C_2F_3Cl_3$, and $n - C_7H_{16}$, the $|\Delta\hat{T}|^{2\beta}$ term dominates (a_3 is relatively large and positive), while in many other fluids, such as HD, Ne, N_2 , and CH_4 , the two singular contributions in diameter partially compensate each other (a_3 is small and negative), creating an illusion of rectilinear diameter even close to the critical point. In Fig. 2, the two asymmetry coefficients are plotted against the dimensional density ρ^* defined as $\rho^* = \rho_c(8\xi_0^3)$, where ξ_0 is the amplitude of the correlation length (representing the range of interactions) obtained from the heat-capacity amplitude A_0^+ through the two-scale factor of universality $A_0^+ \rho_c \xi_0^3 = 0.171$ [5(c)]. A general trend in the two sources of asymmetry is clear: The $|\Delta\hat{T}|^{2\beta}$ singularity is a dominant contribution into the singular diameter if the ratio of critical volume per molecule (ρ_c^{-1}) to “interaction volume ($8\xi_0^3$)” is large [17].

Assuming that the relations between physical fields and scaling fields are not affected by fluctuations, we have developed a method to obtain the asymmetry coefficients

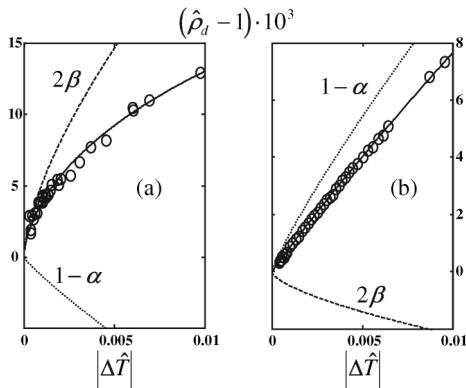


FIG. 1. Singular diameters in (a) SF_6 and (b) N_2 . Experimental data: SF_6 [16(c)] and N_2 [3]. Curves: Solid—fit to Eq. (10); dashed— 2β term; dotted— $1-\alpha$ and linear terms. Heat-capacity source [16(d)] (SF_6) and [16(i)] (N_2).

a_3 and b_2 from mean-field (“classical”) equations of state. By combining Eqs. (3) and (7)–(9), we obtain

$$\frac{a_3}{1+a_3} = \frac{2\mu_{21}}{3\mu_{11}} - \frac{\mu_{40}}{5\mu_{30}}, \quad b_2 = \frac{\mu_{21}}{\mu_{11}^2} - \frac{\mu_{40}}{5\mu_{30}\mu_{11}}, \quad (12)$$

with $\mu_{ij} = \partial^{i+j} \mu / \partial \hat{\rho}^i \partial T^j$. We have obtained a_3 and b_2 for a few classical equations of state, the fine-lattice discretization model (crossover between the van der Waals fluid and lattice gas) [18], the Debye-Hückel and Flory-Huggins models. The coefficient of rectilinear diameter

$$D = \frac{a_3}{1+a_3} \frac{6\mu_{11}}{\mu_{30}} - b_2 \frac{3\mu_{11}^2}{\mu_{30}} = \frac{a_3}{1+a_3} \bar{B}_0^2 - b_2 \frac{\Delta\bar{C}_V}{k_B}, \quad (13)$$

where \bar{B}_0 and $\Delta\bar{C}_V$ are mean-field amplitudes of coexistence densities and heat-capacity discontinuity. Close to the critical point, the rectilinear diameter, affected by fluctuations, splits into two nonanalytical terms, shifting the critical density. In Fig. 3, crossover between rectilinear diameter and complete-scaling singular diameter is shown for the van der Waals equation of state renormalized by fluctuations [19]. A fluctuation shift in the van der Waals critical density is controlled mainly by the $|\Delta\hat{T}|^{2\beta}$ singularity since the van der Waals value of a_3 is relatively large [$a_3/(1+a_3) = b_2/\hat{\mu}_{11} = 0.2$]. In the same fashion, we have calculated crossover between the discontinuity $\Delta(d^2\hat{\mu}/d\hat{T}^2)_{cxc} = (\mu_{11}^2/\mu_{30})[(-\mu_{21}/\mu_{11}) + (3\mu_{40}/5\mu_{30})] = -a_3/(1+a_3)(\Delta\bar{C}_V/k_B)$ and a divergence known as the Yang-Yang anomaly $(d^2\hat{\mu}/d\hat{T}^2)_{cxc} = a_3(d^2\hat{P}/d\hat{T}^2)_{cxc} = -a_3/(1+a_3)A_0^-|\Delta\hat{T}|^{-\alpha}$. A renormalization-group treatment of the $(\Delta\hat{\rho})^5$ term instead pre-

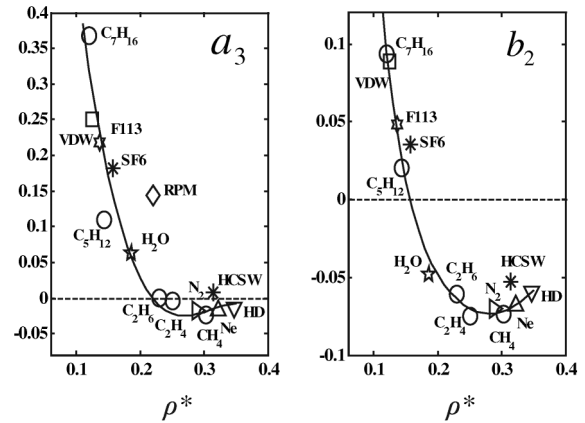


FIG. 2. Complete-scaling asymmetry coefficients a_3 and b_2 vs reduced critical density $\rho^* = \rho_c(8\xi_0^3)$. VDW is a modified-by-fluctuations van der Waals fluid [19] with a short interaction range [$R = (\rho^*)^{1/3} = 0.5$]. HCSW is a simulated hard core square well model [13]. For a simulated restrictive primitive model (RPM) [13] $a_3 = 0.14$ and $b_2 = -0.48$ (off scale) with $\rho^* \approx 0.22$. The solid curves are given as a guide to the eye.

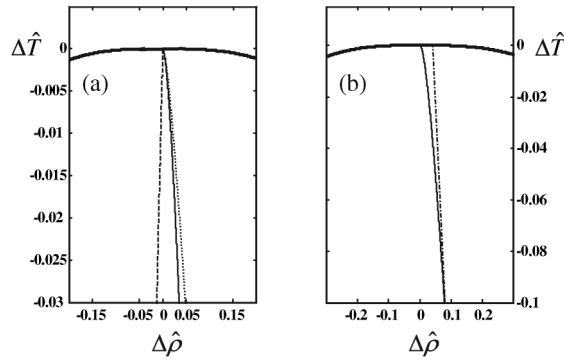


FIG. 3. Crossover diameter in a van der Waals equation of state modified by fluctuations with a short interaction range $R = (\rho^*)^{1/3} = 0.5$. The thick solid curves are the phase boundary. (a) Two contributions in the singular diameter (solid line): $1 - \alpha$ and linear term (dashed line) and 2β (dotted line); (b) crossover between rectilinear diameter (dashed-dotted line) and singular diameter (solid line) in a broader critical region.

dicts a cusp, containing a term $\propto |\Delta\hat{T}|^{\theta_5 - \alpha - \beta} \sim |\Delta\hat{T}|^{0.865}$ [10]. Similarly, there should be a term $\propto a_3/(1 + a_3)|\Delta\hat{T}|^{\beta - \nu}$ instead of $|\Delta\hat{T}|^{\theta_5 - \nu}$ [20] in the so-called Tolman's length, a curvature correction to the surface tension of a liquid droplet.

We conclude that the asymmetry in near-critical fluids originates from two sources: One is a coupling between the density and density of entropy; another one is a nonlinear coupling between the density and molecular volume. Both sources can be incorporated into symmetric Ising criticality by a proper mixing of physical fields into scaling fields.

We acknowledge valuable discussions with M. E. Fisher, Y. C. Kim, C. A. Cerdeiriña, J. V. Sengers, and B. Widom. We also thank I. M. Abdulagatov, E. E. Gorodetskiĭ, and V. P. Voronov for providing us with unpublished heat-capacity data for hydrocarbons. The research was supported by NASA, Grant No. NAG32901.

*Electronic address: anisimov@umd.edu

- [1] L. Cailletet and E. C. Mathias, C.R. Hebd. Seances Acad. Sci. (Paris) **102**, 1202 (1886).
- [2] I. Hahn, M. Weilert, F. Zhong, and M. Barmatz, J. Low Temp. Phys. **137**, 579 (2004).
- [3] M. W. Pestak, M. H. W. Chan, J. R. deBruyn, D. A. Balzarini, and N. W. Ashcroft, Phys. Rev. B **36**, 599 (1987).
- [4] R. R. Singh and K. S. Pitzer, J. Chem. Phys. **92**, 3096 (1990).
- [5] (a) M. E. Fisher, in *Critical Phenomena*, edited by F. J. W. Hahne, Lecture Notes in Physics Vol. 186 (Springer, Berlin, 1982), p. 1; (b) R. Guida and J. Zinn-Justin, J. Phys. A **31**, 8103 (1998); (c) M. E. Fisher and S.-Y. Zinn, J. Phys. A **31**, L629 (1998).
- [6] C. N. Yang and C. P. Yang, Phys. Rev. Lett. **13**, 303 (1964).
- [7] N. D. Mermin and J. J. Rehr, Phys. Rev. Lett. **26**, 1155 (1971); V. L. Pokrovskiĭ, JETP Lett. **17**, 156 (1973); N. B. Wilding and A. D. Bruce, J. Phys. Condens. Matter **4**, 3087 (1992).
- [8] B. Widom, in *Proceedings of The R. A. Welch Foundation Conferences on Chemical Research XVI: Theoretical Chemistry, Houston, 1972* (R. A. Welch Foundation, Houston, 1972).
- [9] U. Narger and D. A. Balzarini, Phys. Rev. B **42**, 6651 (1990).
- [10] M. Ley-Koo and M. S. Green, Phys. Rev. A **23**, 2650 (1981); J. F. Nicoll and R. K. P. Zia, Phys. Rev. B **23**, 6157 (1981); F. Zhang and R. K. P. Zia, J. Phys. A **15**, 3303 (1982); K. E. Newman and E. K. Riedel, Phys. Rev. B **30**, 6615 (1984).
- [11] F. C. Zhang, Ph.D. thesis, Virginia Polytechnic Institute and State University, 1983.
- [12] M. E. Fisher and G. Orkoulas, Phys. Rev. Lett. **85**, 696 (2000); Y. C. Kim, M. E. Fisher, and G. Orkoulas, Phys. Rev. E **67**, 061506 (2003).
- [13] Y. C. Kim, M. E. Fisher, and E. Luijten, Phys. Rev. Lett. **91**, 065701 (2003); Y. C. Kim and M. E. Fisher, Chem. Phys. Lett. **414**, 185 (2005).
- [14] A. K. Wyczalkowska, Y. C. Kim, M. A. Anisimov, and J. V. Sengers, J. Chem. Phys. **116**, 4202 (2002).
- [15] C. Bagnuls and C. Bervillier, Phys. Rev. B **32**, 7209 (1985).
- [16] (a) (C_5H_{12} , cxc) L. M. Artyukhovskaya, E. T. Shimanskaya, and Yu. I. Shimanskiĭ, Sov. Phys. JETP **32**, 375 (1971); (b) ($C_2F_3Cl_3$, cxc) E. T. Shimanskaya, I. V. Bezruchko, V. I. Basok, and Yu. I. Shimanskiĭ, Sov. Phys. JETP **53**, 139 (1981); (c) (SF_6 , cxc) J. Weiner, K. H. Langley, and N. C. Ford, Jr., Phys. Rev. Lett. **32**, 879 (1974); (d) (SF_6 , C_V) A. Haupt and J. Straub, Phys. Rev. E **59**, 1795 (1999); (e) (CH_4 , cxc) R. Kleinrahn and W. Wagner, J. Chem. Thermodyn. **18**, 739 (1986); (f) (H_2O , cxc and C_V) W. Wagner, and A. Pruß, J. Phys. Chem. Ref. Data **31**, 387 (2002); (g) (C_7H_{16} , cxc) L. M. Artyukhovskaya, E. T. Shimanskaya, and Yu. I. Shimanskiĭ, Sov. Phys. JETP **36**, 1140 (1973); (h) (RPM, C_V) G. Orkoulas, M. E. Fisher, and A. Z. Panagiotopoulos, Phys. Rev. E **63**, 051507 (2001); (i) (N_2 and Ar, C_V) A. V. Voronel, V. G. Gorbunova, V. A. Smirnov, N. G. Shmakov, and V. V. Shekochikhina, Sov. Phys. JETP **63**, 965 (1972); (j) (RPM, cxc; HCSW, cxc and C_V) Y. C. Kim, Phys. Rev. E **71**, 051501 (2005).
- [17] This is supported by a recent simulation [R. L. C. Vink, J. Chem. Phys. **124**, 094502 (2006)] of the Widom-Rowlinson model [9] for which $\rho_c \sigma^3 \approx 0.75$ (σ is the particle diameter) and no 2β term is detected.
- [18] S. Moghaddam, Y. C. Kim, and M. E. Fisher, J. Phys. Chem. B **109**, 6824 (2005).
- [19] A. K. Wyczalkowska, J. V. Sengers, and M. A. Anisimov, Physica (Amsterdam) **334A**, 482 (2004).
- [20] M. P. A. Fisher and M. Wortis, Phys. Rev. B **29**, 6252 (1984).