ARTICLES

Surface tension in the Widom model by low-temperature expansion

Arjun Berera*
Department of Physics, University of California, Berkeley, Berkeley, California 94720

Byungnam Kahng
Department of Physics, Kun Kook University, Seoul 133, Korea
(Received 20 December 1991; revised manuscript received 11 May 1992)

The surface tension between coexisting plus and minus ferromagnetic phases of the Widom model [J. Chem. Phys. **84**, 6943 (1986)] is computed by performing a seventh-order low-temperature expansion. A functional form for the surface tension is suggested, based on Padé-approximant analysis, for the complete range from $T=0$ to bulk criticality. The surface tension is computed along the roughening line, which had been recently calculated by a low-temperature expansion [B. Kahng, A. Berera, and K. A. Dawson, Phys. Rev. A **42**, 6093 (1990)]. The positivity and well-behaved form of our surface-tension series along this line establishes the validity of the low-temperature approximation in this regime of parameter space. Comparisons of the low-temperature results to mean-field theory are made. A preliminary examination of the critical amplitude is discussed.

PACS number(s): 05.50.+q

I. INTRODUCTION

At a temperature of about one-half bulk criticality, it has long been known that a three-dimensional nearest-neighbor Ising model with a plus-minus interface will undergo a transition from a low-temperature "flat-surface" phase to one in which the average interfacial width diverges. The onset of the latter phase is called the roughening transition.

An interesting characteristic of the rough phase is the presence of long-range correlations at all temperatures above the roughening transition temperature T_R to the bulk critical point T_c. A simple physical picture for such a transition is to imagine that for low enough temperatures the interface effectively behaves like a two-dimensional nearest-neighbor Ising model and that the roughening transition corresponds to the usual order-disorder transition in the 2D Ising model. Arguments based on spin-flip energetics qualitatively justify this picture near the transition point because of the fact that the interactions of the interfacial spins are of opposite signs in the two adjacent layers next to an interfacial layer. However, the persistence of the roughened phase above T_R is indicative of a more complex mechanism. It is precisely this complexity that results in producing a rich set of phenomena that have much interest both for their physical and theoretical implications.

In the present work, we will examine the roughening transition in the Widom model of microemulsions [1], a three-dimensional spatially isotropic, frustrated lattice model, with Hamiltonian

$$H=-\frac{J}{2}\sum_{n,n'}^{\mathrm{NN}}\sigma_n\sigma_{n'}-\frac{\gamma M}{2}\sum_{n,n'}^{\mathrm{DNN}}\sigma_n\sigma_{n'}-\frac{M}{2}\sum_{n,n'}^{\mathrm{LNNN}}\sigma_n\sigma_{n'}\,, \tag{1.1}$$

where the sum in the first term is over the six nearest neighbors, in the second term over the twelve diagonal nearest neighbors, and in the last term over the six linear next-nearest neighbors. We will only examine the case where $\gamma=2$. For this case the phase diagram, obtained from Monte Carlo simulation [2], is shown in Fig. 1, where in reference to the figure $j\equiv J/kT$ and $m\equiv M/kT$.

This model is a useful paradigm for a broad class of bulk and surface phenomena not described by the nearest-neighbor Ising model. Throughout most of the bulk low-temperature phase diagram one finds stable ordered structures that disorder via fluctuation-induced first-order phase transitions. There are a number of multiphase points such as R, P_2, and Q_2 in Fig. 1, from which the low-temperature phases originate to form a manifold of ordered structures with one, two, and three-dimensional orders. In the vicinity of these multiphase points, the interfacial tension between any two phases becomes very small, vanishing at the point itself. The interfacial tensions are thus extremely low near zero and the critical temperatures, an aspect that is absent in unfrustrated systems. Since this is one of the simplest models that exhibits these phenomena and the Hamiltonian has the attractive feature of being isotropic, it shows potential applicability not just for microemulsions but also for alloy systems and studies of magnetic order.

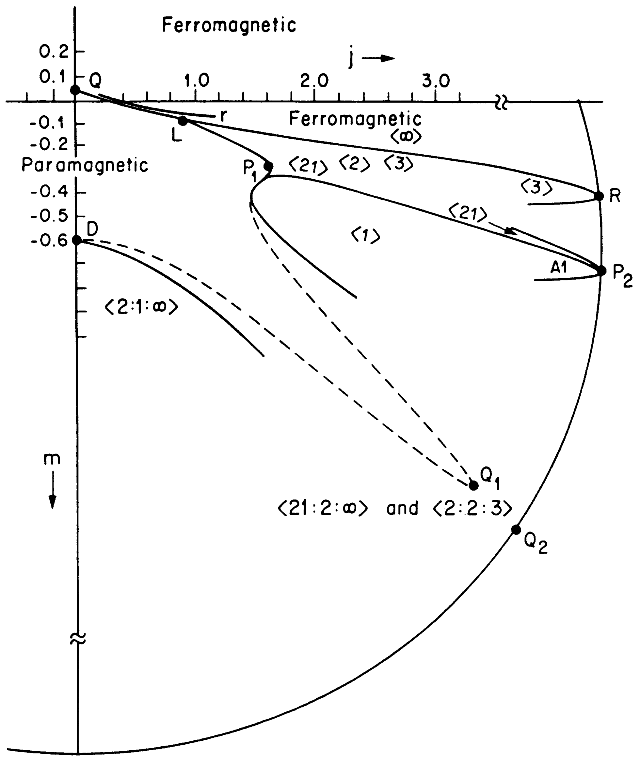

FIG. 1. Monte Carlo phase diagram obtained from Ref. [2] for the Widom model at $\gamma=2$. A sketch of the roughening line obtained in Ref. [4] is denoted by r.

We have been pursuing a series of studies of the low-temperature behavior [3,4] of the Widom model. One of the purposes of this communication is to complete the study in Ref. [4], which examined the roughening transition in the ferromagnetic two-phase region. To briefly summarize the relevant aspects of that paper, it was found that a line of roughening transitions existed in the j-m plane. This conclusion was based on studying several different seventh-order low-temperature series expansions. Each of the series were chosen so that in exact form they would exhibit the roughening singularity if one existed. A schematic picture of the roughening line that was obtained from the analysis in Ref. [4] and its relation to the rest of the phase diagram is shown as line r in Fig. 1.

Due to the nature of this, as most low-temperature expansions, there is no *a priori* way of knowing its region of validity. That is to say, without knowledge of the higher-order terms, one is generally forced to rely on how reasonable the results are qualitatively and then decide on their quantitative validity. In the present case of the Widom model the problem is accentuated since there is little in the way of a developed intuition, as yet, about this model. For the case in Ref. [4], since we were able to find the nontrivial singular behavior associated with roughening, we felt confident that our expansion was valid. However, finding the singularity is not sufficient because one must also know that it occurs below the temperature where, to the same order of approximation, the surface tension is still positive. Otherwise the singularity we found would have to be interpreted as just some artifact of a low-order series. As such a more stringent test would be to compute the surface tension along the seventh-order approximated roughening line and make sure it is positive. This will establish complete internal consistency of the low-temperature expansion out to seventh order. With this as our primary goal, in this paper we present the low-temperature expansion series to seventh order for the surface tension $\Sigma(T)$ between coexisting plus and minus ferromagnetic phases, study this series numerically, and confirm positive surface tension along the roughening line.

The paper is organized as follows. The low-temperature expansion of the surface tension and its evaluation along the roughening line is given in Sec. II. Then in Sec. III, comparisons of our series with mean-field theory will be made. Finally in Sec. IV, following the work of Shaw and Fisher [5], a preliminary examination is made of the surface-tension critical amplitude A_Σ defined by the behavior of the surface tension at the bulk critical point as

$$\Sigma(t) \approx A_\Sigma t^\mu , \tag{1.2}$$

where $t \equiv |T-T_c|/T_c$ and μ is the interfacial tension critical exponent. However, due to insufficient accuracy in the available estimates of the roughening critical temperatures, we are unable at present to make reliable quantitative statements about A_Σ. We feel that more comprehensive studies of our series in Ref. IV are necessary before further calculations in A_Σ can be attempted.

II. SURFACE TENSION

In this section we construct the low-temperature series for Σ and evaluate it along the roughening curve. The coefficients of $\Sigma(y)$ that we have obtained are given in the Appendix, Eq. (A1). There, two expansion coefficients are used, the small parameter

$$y = e^{-4J/kT} \tag{2.1}$$

and the auxiliary parameter

$$x = e^{-4M/kT} . \tag{2.2}$$

Note that the latter is not meant to be small necessarily. It is introduced merely as a notational convenience.

To compute the coefficients, a modified Martin-type algorithm [6] was used to obtain all connected clusters and clusters with only one disconnected monomer. The remaining clusters were computed by hand. Our algorithm incorporates summing over nearest, next-nearest, and diagonal neighbor interactions and thus is more elaborate than the original Martin algorithm for the nearest-neighbor Ising model. The extension to even higher orders in the low-temperature expansion is mainly hindered by the difficulty of counting the terms done by hand. Observe that in the limit $M=0.0$, or equivalently $x=1$, (A1) is the series for the surface tension of the three-dimensional Ising model [7].

Our numerical work is based on both the direct evaluation of the series and a Padé approximant technique. For the latter, in analogy to the work of Shaw and Fisher [5], we define the function

$$Q_x(y) \equiv x^5 y^{1/2} \exp\left[\frac{\Sigma(y)}{kT}\right] . \tag{2.3}$$

This definition is convenient since it removes the zero-temperature limit energy term, hence permitting a Padé approximant in the parameter y. We emphasize that in this definition we do not attempt to well represent the nonanalyticity at the roughening transition. Rather, we seek an approximant to the interfacial tension below T_R, the roughening critical temperature, which extrapolates the low-order low-temperature series.

We expect Padé approximants of the low-temperature series represented through $Q_x(y)$ to converge well up to the first singularity, the roughening temperature. Beyond this we extend the fit by using a suitable approximant [5,8], which matches with Σ and its derivatives at T_R and vanishes at T_c. The general form we consider is

$$\Sigma_{[N,D]}(t) = t^{\mu} P_{[N,D]}(t) . \tag{2.4}$$

This equation can be taken as the definition of $P_{[N,D]}(t)$. To construct the Padé approximant, we expand the known low-temperature series for Σ in terms of the variable $t - t_R$, obtain the desired $[N,D]$ approximant, and then reexpress the result in terms of the variable t. A particular low-order approximant $P_{[0,1]}$, valid in the range $T_R < T < T_c$ is

$$P_{[0,1]}(t) = \frac{C}{1+Dt} , \tag{2.5}$$

where

$$C = \frac{C_1}{1 - D_1 t_R} , \tag{2.6}$$

$$D = \frac{D_1}{1 - D_1 t_R} , \tag{2.7}$$

and

$$C_1 = t_R^{-\mu} \Sigma_R^{(0)} , \tag{2.8}$$

$$D_1 = \frac{\mu t_R^{-(\mu+1)} \Sigma_R^{(0)} - t_R^{-\mu} \Sigma_R^{(1)}}{C_1} , \tag{2.9}$$

with $\Sigma_R^{(0)} \equiv \Sigma(t_R)$, $\Sigma_R^{(1)} \equiv d\Sigma/dt(t_R)$, and $t_R \equiv (T_c - T_R)/T_c$.

Hence for any desired value of x, one can use $P_{[0,1]}$ as given in (2.5) and a suitable Padé approximant to $Q_x(y)$ defined in (2.3) to obtain a closed-form expression of Σ

TABLE I. Low-temperature expansion results for the surface tension along the roughening line by three different series approximation approaches. The data for the roughening line were taken from Ref. [4] based on the two series (using their denotations of these series) (a) $\langle z^2 \rangle_{BO}$ and (b) R'_{BO}. The asterisk denotes spurious poles encountered.

		$\frac{\Sigma}{2J+20M}$		
m_R	j_R	Direct	$Q_x(y)$ [7,0]—approximate	$Q_x(y)$—averaged
		(a) $\langle z^2 \rangle_{BO}$		
0.0	0.385	0.804	0.810	0.787
−0.01	0.439	0.811	0.817	0.796
−0.02	0.495	0.823	0.828	0.803
−0.03	0.555	0.842	0.846	0.819
−0.04	0.619	0.869	0.871	0.849
−0.05	0.691	0.902	0.903	0.889
−0.06	0.771	0.936	0.936	0.930
−0.07	0.863	0.965	0.965	0.963
−0.08	0.976	0.985	0.985	0.985
−0.09	1.132	0.996	0.996	0.996
		(b) R'_{BO}		
0.0	0.359	0.714	0.729	0.668
−0.01	0.450	0.839	0.843	0.829
−0.02	0.514	0.867	0.869	0.858
−0.03	0.569	0.874	0.876	0.862
−0.04	0.621	0.873	0.875	0.855
−0.05	0.672	0.864	0.866	0.830
−0.06	0.721	0.841	0.843	0.747
−0.07	0.769	0.778	0.781	*
−0.08	0.815	0.272	0.281	0.486
−0.09	0.860	1.155	1.152	1.172
−0.10	0.903	0.992	0.990	1.028
−0.11	0.944	0.923	0.918	0.956
−0.12	0.982	0.845	0.830	*

for the complete range $0 < T < T_c$. At present, the uncertainties in the known values of both T_R and T_c make the complete program, for general x, too ambitious.

We now turn to the results for Σ, given in Table I, which were evaluated at values on the roughening lines. The data for the roughening lines were obtained from Ref. [4] from two different series denoted [see Eqs. (2.1) to (2.5) of that paper] $\langle z^2 \rangle_{\rm BO}$ and $R'_{\rm BO}$ corresponding to the cases of parts (a) and (b) of Table I, respectively (the index BO means that the series includes bulk excitations and overhangs). We clarify that we expect only one "true" roughening line on the phase diagram. However, as stated in the Introduction, we examined several series in Ref. [4] to determine this roughening line, each giving approximately but not exactly the same line. In the present analysis we used the results from the two above-named series, since they were the most relevant. In Table I, the first two columns give the coordinates of the roughening line in the j-m plane. Column 3 gives the direct evaluation of the series, column 4 is based on an expansion of $Q_x(y)$ to seventh order, and column 5 is based on averaging the [2,5], [3,4], [4,3], [5,2] Padé approximants of $Q_x(y)$. This choice for the average was made simply to give a concise summary of the results for the near diagonal Padé approximants.

These results confirm positivity of the surface tension and so as discussed earlier demonstrate the consistency of our expansions in [4]. Furthermore, from Table I one sees that the three ways we used to calculate Σ give approximately the same results within each of the parts of the table and also in comparing the two parts. Some of the entries in the fifth column, based on the average, do show significant differences from columns 3 and 4. This is a consequence of variations between the $[L,M]$ Padé approximants used in our average. The most striking feature is in part (b) of Table I, where Σ suddenly dips down at $m=-0.08$, jumps up at $m=-0.09$, and then steadily decreases again. This is not an effect of a low-order series, but arises in fact because the ratio $|j/m|$ becomes less than 10.0, which in the zero-temperature phase diagram is outside of the ferromagnetic phase and in the region of the layered phase.

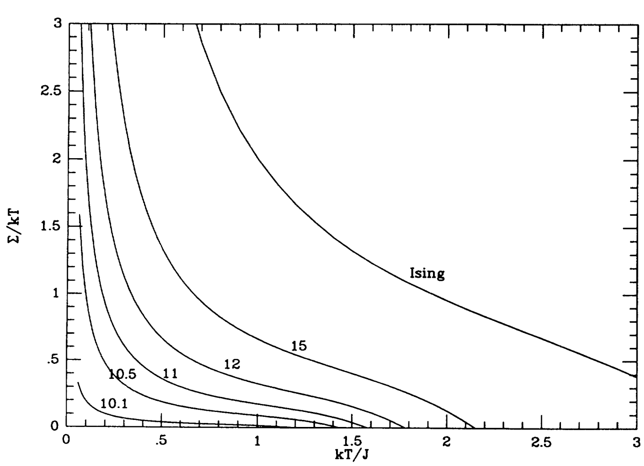

FIG. 2. Surface tension from the direct evaluation of series for Ising limit (top) and $j+km=0$ with $k=15,12,11,10.5,10.1$.

TABLE II. Estimated coordinates of bulk critical points based on series for surface tension.

m	j
0.00	0.29
−0.01	0.34
−0.02	0.40
−0.05	0.58
−0.10	0.76

A general survey of the behavior of Σ is displayed in Fig. 2 for a wide parameter range. The surface tensions in this figure were obtained by a direct evaluation of our series, without the use of any Padé approximants. Note that the normalization of Σ amounts to a division only by kT. The graphs of Σ/kT are for the values of $j+km=0$, with $k=15.0, 12.0, 11.0, 10.5, 10.1$ and also the Ising limit, which is the uppermost curve.

An interesting feature of the present model occurs at $j+10m=0$, which for one thing is a point of degeneracy when $T=0$. In this limit it turns out that the surface tension vanishes at $T=0$, which is in addition to the usually expected case of $T\to\infty$. This is demonstrated in Fig. 2 by the fact that Σ decreases in the double limit $k\to 10, T\to 0$.

It is also possible to estimate the bulk transition temperature T_c from the ferromagnetic to the disordered paramagnetic phase by computing where Σ vanishes. These estimates have to be taken with the caveat that Σ has a nonanalytic term at T_R, although Σ and all its derivatives are smooth across T_R. Table II presents some estimated critical temperatures in terms of the coordinates j and m along the critical-point locus. They were obtained by approaching along the j direction while holding m fixed at various values.

III. COMPARISON TO MEAN-FIELD THEORY

In Fig. 3 a comparison between the low-temperature series (solid lines) and mean-field theory [9] (dashed lines) is shown. We adhere to the same choice of coordinate axis as used in Ref. [9] so as to make the most direct comparison. The curves exhibit the surface tension at the fixed values of $1/(j+3m)=1.0$ (top) and 2.5 (bottom). The curves from the low-temperature series were obtained by the direct evaluation of the series. As an aside, we remark that the comparison of the low-temperature data between the direct evaluation of the series, as shown, and the averaged Padé approximant analysis, as defined for column five of Table I, is very good for the top curve and shows marked disagreement for the bottom curve.

Turning to the comparison with mean-field theory, we see that the agreement in Fig. 3 is excellent for the case $\beta\equiv 1/(j+3m)=1.0$ and poor for $1/(j+3m)=2.5$, where there is total breakdown. The former is a good parameter regime for both methods and so the good comparison is understandable. To understand the breakdown

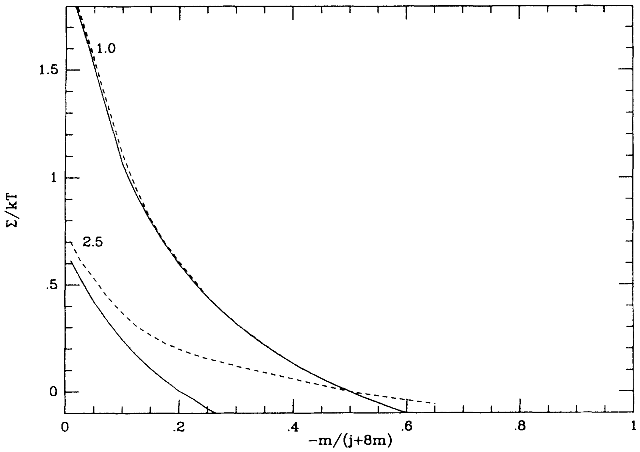

FIG. 3. Comparison of direct series evaluation of surface tension (solid line) to mean-field theory results (dashed lines) for $1/(j+3m)=1.0$ and 2.5.

in the latter, it is beneficial to note what the corresponding ratio j/m and the magnitudes of j and m are in relation to the coordinates in Fig. 3. In these terms, as $\alpha \equiv -m/(j+8m)$ goes from 0 to 1, the ratio j/m goes from $-\infty$ to -9 with, in particular, $j/m=-10.0$ at $-m/(j+8m)=0.5$. For fixed value along the abscissa $[-m/(j+8m)=\text{const}]$, as one moves from $1/(j+3m)=1.0$ to $1/(j+3m)=2.5$, the magnitudes of j and m decrease. To be precise, the relation between j and m to α and β is

$$j=\frac{1}{\beta\left[1-\dfrac{3\alpha}{1+8\alpha}\right]}, \tag{3.1}$$

$$m=\frac{-\alpha}{\beta(1+5\alpha)}. \tag{3.2}$$

In the context of the phase diagram in Fig. 1, going from $\beta=1.0$ to $\beta=2.5$ at fixed α corresponds to moving along a ray of fixed ratio j/m toward increasing temperature.

Higher temperatures immediately imply that the low-temperature series is less reliable. In the region of interest here, one can establish that the low-temperature series is at the onset of breaking down. For this an estimate at $\beta=2.5$ and $\alpha=0$ shows that the fifth-, sixth-, and seventh-order terms from the series (A1) are all at the same magnitude of about 0.01. Similar behavior is found all along the $\beta=2.5$ line at higher α. This lining up of successive terms is indicative of a breakdown of the series.

Turning to the mean-field theory curve, we cannot speak for the results of [9], but one observation is noteworthy. Since the parameter region under discussion corresponds to the ferromagnetic phase, not the paramagnetic phase, the corresponding temperatures are, relatively speaking, intermediate, not high. In this regime it is well known that mean-field theory is not ideal. However, unlike for the low-temperature series, it is hard to assess within the confines of mean-field theory its precise region of validity. Beyond the above remarks, we can only say that the best method to understand this region, should it be of interest, is Monte Carlo simulation.

IV. ISSUE OF CRITICAL AMPLITUDE

Based on the approximant given in Eq. (2.5), which extrapolates Σ from T_R up to T_c, one can compute the critical amplitude easily by inspection to be $A_\Sigma=C$, where C is given in Eq. (2.6). Unfortunately, we have found from our numerical studies that A_Σ is highly sensitive to the value of T_R that one uses. In particular, the derivatives of Σ vary drastically for small changes in T_R. As such, we do not quote any estimates of A_Σ at this stage. A more careful analysis of our series in Ref. [4], using more reliable approximants, seems necessary before further progress can be made. Nevertheless, given this data, estimates of A_Σ can be obtained directly using Eq. (2.4) to (2.9).

V. CONCLUSIONS

In this paper we have presented a closed formula that describes the variation of the interfacial tension throughout the ferromagnetic region of the phase diagram fairly well. It should be quite useful for a number of applications including, for example, a description of the interfacial tension of amphiphilic mixtures as a function of amphiphile concentration [1,9], and nucleation and growth studies of magnetic domains where the exchange interaction causes frustration.

Nevertheless, our quantitative knowledge of the Widom model lags far behind that of the much simpler nearest-neighbor Ising model. In particular, further progress could be achieved by deeper analysis of the various low-temperature series, which have already been obtained for this model.

ACKNOWLEDGMENTS

Arjun Berera thanks Professor Michael E. Fisher and Dr. Laurence J. Shaw for helpful conversations. We also acknowledge discussion with Professor Kenneth A. Dawson and thank him for use of his computer facilities. Funding was provided to A. B. by the Department of Education and to B. K. from the Regents of the University of California. Partial funding was also provided by the Sloan Foundation.

APPENDIX

The low-temperature series for the surface tension Σ is

$$\frac{\Sigma}{kT}=2j+20m-(y^2[2x^6]+y^3[4x^2+2x^{14}-4x^{15}]+y^4[4x^{10}+4x^{11}-18x^{12}+4x^{14}+8x^{16}+4x^{17}+4x^{20}]$$
$$+y^5[x^{12}+16x^{16}+24x^{17}-96x^{18}+8x^{19}+22x^{20}+16x^{21}+4x^{22}+16x^{23}+8x^{24}+4x^{26}$$
$$+4x^{28}+2x^{29}-13x^{30}]+y^6[8x^{13}+4x^{14}+16x^{15}-92x^{16}-120x^{17}+\tfrac{848}{3}x^{18}$$
$$+40x^{19}-62x^{20}+68x^{21}-196x^{22}-92x^{23}-4x^{24}$$
$$+80x^{25}-28x^{26}+44x^{27}+25x^{28}+14x^{29}$$
$$+73x^{30}+14x^{32}+2x^{34}+4x^{36}]$$
$$+y^7[8x^{16}+8x^{17}-26x^{18}+40x^{19}+40x^{20}+148x^{21}-660x^{22}-1160x^{23}+2732x^{24}$$
$$+100x^{25}-852x^{26}-194x^{27}-404x^{28}-632x^{29}+160x^{30}+532x^{31}-196x^{32}$$
$$+272x^{33}+94x^{34}+60x^{35}+72x^{36}+32x^{38}+8x^{39}+16x^{40}+12x^{41}+8x^{42}$$
$$-54x^{43}-14x^{44}])\ . \tag{A1}$$

The low-temperature series for the free energy per site for the bulk ferromagnetic phase in the Widom model is

$$\frac{F}{kT}=3j+15m-(y^3[x^{15}]+y^5[3x^{30}]+y^6[6x^{28}+3x^{29}-\tfrac{25}{2}x^{30}]+y^7[12x^{43}+3x^{44}])\ . \tag{A2}$$

---

*Present address: Department of Physics, University of Arizona, Tucson, AZ 85721.

[1] B. Widom, J. Chem. Phys. **84**, 6943 (1986).

[2] K. A. Dawson, B. Walker, and A. Berera, Physica A **165**, 320 (1990).

[3] A. Berera and K. A. Dawson, Phys. Rev. A **41**, 626 (1990); **42**, 3618 (1990).

[4] B. Kahng, A. Berera, and K. A. Dawson, Phys. Rev. A **42**, 6093 (1990).

[5] L. J. Shaw and M. E. Fisher, Phys. Rev. A **39**, 2189 (1989).

[6] J. L. Martin, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1972), Vol. 3; S. Redner, J. Stat. Phys. **29**, 309 (1982).

[7] Please see Ref. [1] cited in Ref. [5] for the series of the surface tension in the three-dimensional Ising model, which was obtained by J. D. Weeks, G. H. Gilmer, and H. J. Leamy.

[8] In Ref. [5] we find a discrepancy of a minus sign between their definition of $(d\Sigma/dt)|_{T_R}$ and the values quoted in their Table I. Recalling their definition of t for the region $0<T<T_c, t\equiv(T_c-T)/T_c$, it follows that $(d\Sigma/dt)|_{T_R} = [(dT/dt)(d\Sigma/dT)]|_{T_R} = [(-1/T_c)(d\Sigma/dT)]|_{T_R}>0$. Therefore it seems the results of Table I, column 3, of that paper do not need the minus signs which are present. This is also then consistent with their results in Sec. III.

[9] K. A. Dawson, Phys. Rev. A **35**, 1766 (1987).