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# Universal relations among thermodynamic critical amplitudes

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The hypothesis of universality implies that there are four universal ratios among the six usually defined thermodynamic critical amplitudes. Theoretical information from series and $\epsilon$ expansions is presented on the values of these ratios for short-ranged Ising, Heisenberg, and spherical models, and for dipolar systems. A number of real materials are discussed (Xe, $CO_2$, Ni, EuO, and $LiTbF_4$), and the present state of our understanding of the thermodynamic ratios for these systems is found to be rather crude.

## I. INTRODUCTION

The phenomenological theories of scaling[1,2] and universality[3] have contributed significantly to our understanding of the thermodynamic properties of real materials near their critical point.[4,5] In practice, experimental data are often represented in terms of scaled equations of state, whose exponents are constrained to satisfy the scaling relations. Even when independent measurements of the exponents are made, the scaling laws are tested, and the values of the exponents are used to designate universality classes. It has become clear in recent years,[5-8] however, that ratios of *critical amplitudes* play an equally important role in characterizing universality classes, and that a significant amount of information is available if amplitudes are also properly analyzed.

The renormalization-group theory of critical phenomena[9] has deepened our understanding of scaling and universality, and has provided a number of calculational tools for estimating universal properties, i.e., both exponents and amplitude ratios. The purpose of the present paper is to collect and complete the presently known theoretical information concerning thermodynamic amplitude ratios for different universality classes. We shall draw on the results of series expansions, and also on the $\epsilon$-expansion method using the renormalization group. Most of the results we present have appeared, either implicitly or explicitly, in earlier work, but we believe that it is useful to give a unified presentation of the amplitude ratios, in order to focus the attention of experimentalists on their importance. Our brief consideration of measured values of the ratios in Sec. IV demonstrates that little reliable information exists at the present time on their systematics for different classes of real materials.

Although the $\epsilon$ expansion is not expected to be particularly reliable for three dimensions, it is a versatile method, which can be applied to a wide range of models, some of which are inaccessible to series expansions. An example is the low-temperature specific heat of the Heisenberg model. Another advantage of the $\epsilon$ expansion is that it makes various trends, such as the $n$ dependence of the amplitudes ($n$ is the degree of symmetry of the order parameter), quite apparent, and suggests which ratios will vary smoothly as a function of $n$. It would of course be highly desirable to develop approximate methods for calculating amplitudes using the renormalization group in three dimensions,[10] but until such techniques become available, we believe that the present combination of series and $\epsilon$ expansions gives a rather clear picture of the values of the universal amplitude ratios.

In Sec. II the notation for universal and nonuniversal quantities is defined, and various thermodynamic amplitude ratios introduced. Section III gives the $\epsilon$ expansion of the ratios for short-range models, which follow from the second-order scaled equation of state.[11,12] New results are presented for dipolar systems, for both $n=1$, $d=3$, and $n=d=4-\epsilon$. Series expansions are considered for both Ising and Heisenberg systems in Sec. IV, as well as the results of scaled-equation-of-state analyses of some fluids and magnets.[4,5] Numerical estimates for the amplitude ratios are collected in the tables, and a number of calculations on model systems are in the appendixes.

## II. UNIVERSAL AND NONUNIVERSAL QUANTITIES

In this section we shall introduce our notation for the various universal and nonuniversal thermodynamic quantities. Unless otherwise noted, we shall follow the notation of Ref. 2 and 5, but we shall also introduce a number of new symbols for the critical amplitude ratios. Using the magnetic language, the equation of state near the critical point may be written[2]

$$H = M|M|^{\delta-1}h(x), \tag{2.1}$$

where

$$x = t|M|^{-1/\beta}, \tag{2.2}$$

$t = (T - T_c)/T_c$, $H$ is the magnetic field (or the chemical potential $\mu - \mu_c$), $M$ the magnetization (density $\rho - \rho_c$), and $T$ the temperature. It is often convenient to assume that $H$ and $M$ are already given in dimensionless units[5]; i.e., $H$ is measured in units of

$$H_N = k_B T_c / S g \mu_B \tag{2.3a}$$

($S$ is the spin, $g$ the $g$ factor, and $\mu_B$ the Bohr magneton) and $M$ (the magnetization per mole) is measured in units of

$$M_N = N S g \mu_B \tag{2.4a}$$

($N$ is Avogadro's number). The specific heat $C$ (per mole) is measured in units of

$$C_N^{\text{mag}} = H_N M_N / T_c = R = 8.317 \text{ J mole}^{-1}\text{K}^{-1}. \tag{2.5a}$$

For fluids, $\rho$ is in units of

$$\rho_N = \rho_c, \tag{2.3b}$$

and $\mu$ is in units of

$$\mu_N = P_c/\rho_c, \tag{2.4b}$$

while $C$ is in units of

$$C_N^{\text{fl}} = P_c w / T_c \rho_c, \tag{2.5b}$$

where $P_c$ is the critical pressure, $\rho_c$ the critical density, and $w$ the molecular weight. The formulas we shall write down in what follows will hold for dimensioned variables, and also for the (nonuniversal) dimensionless quantities introduced above. (In the latter case one should set $C_N = H_N = M_N = 1$. We shall use the same notation for the dimensioned and dimensionless quantities.)

The relation (2.1) can be used to obtain all the relevant critical amplitudes. Following Ref. 2, these are defined by

$$M = B(-t)^{\beta}, \quad t<0, \quad H=0 \tag{2.6}$$

$$\chi = \Gamma t^{-\gamma}, \quad t>0, \quad H=0 \tag{2.7}$$

$$\chi = \Gamma'(-t)^{-\gamma'}, \quad t<0, \quad H=0 \tag{2.8}$$

$$C_M = C_H = (A/\alpha)t^{-\alpha} + C_B, \quad t>0, \quad H=0 \tag{2.9}$$

$$C_H = (A'/\alpha')(-t)^{-\alpha'} + C_B', \quad t<0, \quad H=0 \tag{2.10}$$

$$H = DM|M|^{\delta-1}, \quad t=0 \tag{2.11}$$

where $\chi$ is the susceptibility, and $\alpha$, $\alpha'$, $\beta$, $\gamma$, $\gamma'$, and $\delta$ are the usual critical exponents, which satisfy the scaling relations[1,2]

$$\alpha = \alpha', \quad \gamma = \gamma', \quad \gamma = \beta(\delta - 1), \quad \alpha = 2 - 2\beta - \delta. \tag{2.12}$$

Note that $A$ and $A'$ are the amplitudes for the *singular parts* of the respective specific heats, and $C_B$ and $C_B'$ are constants. All the expressions are valid asymptotically, for $t, H \to 0$.

We next define the two nonuniversal constants $x_0$ and $h_0$ through

$$h_0 = h(0), \quad h(-x_0) = 0. \tag{2.13}$$

Rescaling $h(x)$ by $h_0$ and $x$ by $x_0$, we finally arrive at the universal equation-of-state scaling function,[5]

$$\tilde{h}(\tilde{x}) = \tilde{h}(x/x_0) = h_0^{-1}h(x). \tag{2.14}$$

The hypothesis of universality[3] states that the function $\tilde{h}(\tilde{x})$ is the same for all systems within a given equivalence class. From Eq. (2.1) one immediately finds that[2,13]

$$B = x_0^{-\beta}, \tag{2.15}$$

$$\Gamma = \lim_{x\to\infty}[x^{\gamma}/h(x)] = x_0^{\gamma} h_0^{-1}\tilde{\Gamma}, \tag{2.16}$$

$$\Gamma' = \beta x_0^{\gamma-1}/h'(-x_0) = x_0^{\gamma} h_0^{-1}\tilde{\Gamma}', \tag{2.17}$$

$$(M_N H_N / C_N)A = \alpha\beta\int_0^{\infty} h''(y)y^{\alpha-1}\,dy = h_0 x_0^{\alpha-2}\tilde{A}, \tag{2.18}$$

$$(M_N H_N / C_N)A' = \alpha\beta\left(\int_{-x_0}^{0} h''(y)|y|^{\alpha-1}\,dy + x_0^{\alpha-1}h'(-x_0)\right) = h_0 x_0^{\alpha-2}\tilde{A}', \tag{2.19}$$

and

$$D = h_0, \tag{2.20}$$

where $\tilde{\Gamma}$, $\tilde{\Gamma}'$, $\tilde{A}$, and $\tilde{A}'$, are given by[13]

$$\tilde{\Gamma} = \lim_{\tilde{x}\to\infty}[\tilde{x}^{\gamma}/\tilde{h}(\tilde{x})], \tag{2.21}$$

$$\tilde{A} = \alpha\beta\int_0^{\infty} \tilde{h}''(y)y^{\alpha-1}\,dy, \tag{2.22}$$

etc. [Note that Eqs. (2.18) and (2.19) are valid for $\alpha > 0$. The case $\alpha < 0$ is mentioned in Appendix B.]

Since $\tilde{h}(\tilde{x})$ is a universal function, it follows that $\tilde{\Gamma}$, $\tilde{\Gamma}'$, $\tilde{A}$, $\tilde{A}'$, $\tilde{B} \equiv 1$, and $\tilde{D} \equiv 1$ are all universal. All the nonuniversal features in the critical amplitudes are reflected through their dependence on the two scales $x_0$ and $h_0$. Any combination of nonuniversal amplitudes which is independent of $x_0$ and $h_0$ is thus universal.

Since there are only two independent scales, there must be four universal relations among the six amplitudes $A$, $A'$, $\Gamma$, $\Gamma'$, $B$, and $D$. These may be chosen in direct correspondence to the scaling relations (2.12), which reduce the six exponents $\alpha$, $\alpha'$, $\gamma$, $\gamma'$, $\beta$, and $\delta$ to two independent ones. We therefore define the four universal ratios

$$A/A' = \tilde{A}/\tilde{A}', \tag{2.23}$$

$$\Gamma/\Gamma' = \tilde{\Gamma}/\tilde{\Gamma}', \tag{2.24}$$

$$R_\chi \equiv \Gamma D B^{\delta-1} = \tilde{\Gamma}, \tag{2.25}$$

and

$$R_C \equiv (M_N H_N / C_N) A B^{-2} \Gamma = \tilde{A}\tilde{\Gamma}. \tag{2.26}$$

In terms of these quantities we may write, for $t \to 0^+$,

$$\chi(t) = R_\chi H^{-1}[M(H=0, -t)]^{1-\delta}[M(H, t=0)]^{\delta}, \tag{2.27}$$

$$(M_N H_N / C_N) C^s(t) = R_C \alpha^{-1} t^{-2} \times [M(H=0, -t)]^2 \chi^{-1}(t), \tag{2.28}$$

$$C^s(-t) = (A/A')^{-1} C^s(t), \tag{2.29}$$

$$\chi(-t) = (\Gamma/\Gamma')^{-1}\chi(t). \tag{2.30}$$

[The superscript of $C^s$ indicates the singular part.] These relations express the thermodynamic functions entirely in terms of universal amplitudes, and of the magnetization along the coexistence curve and critical isochore.

The quantities $R_\chi$ and $A/A'$ were discussed in some detail in Ref. 5. The ratios $R_\chi$ and $R_C$ were considered by Bauer and Brown.[8] In terms of their notation, we have

$$R_\chi = UR, \tag{2.31a}$$

$$R_C = \alpha R. \tag{2.31b}$$

The ratio $R_\chi$ was also considered by Tarko and Fisher,[14] who defined a quantity $Q_1$ given by

$$R_\chi = Q_1^{-\delta}. \tag{2.31c}$$

It may be noted at this point, that since there are three fundamental thermodynamic quantities ($M$, $\chi$, and $C$) and three singled-out paths (the critical isochore for $t>0$ and $t<0$, and the critical isotherm), there are in general nine critical exponents and amplitudes. Since, however, $M$ is identically zero on the critical isochore for $t>0$, and $\chi$ and $M$ are exactly related to each other on the critical isotherm,

$$\chi(t=0, H) = (\delta D)^{-1}|M(t=0, H)|^{1-\delta},$$

one is left with seven exponents and amplitudes. Apart from the six already considered, one can define an exponent $\alpha_c$ and an amplitude $A_c$ for the specific heat on the critical isotherm, with associated scaling laws and amplitude ratios. Since this quantity is rarely measured, we have not evaluated these ratios in this paper. We may also mention that $\Gamma'$ is finite only in the Ising case ($n=1$), so for $n \geq 2$ there are in all six exponents and amplitudes, and four universal relations among them (including $\alpha_c$ and $A_c$).

Let us note finally that the above discussion, and especially the definitions of amplitudes through Eqs. (2.6)–(2.11), must be modified for the three-dimensional dipolar Ising model.[15,16] In that case, one has

$$M = \hat{B}(-t)^{1/2}|\ln t|^{1/3}, \quad t<0, \quad H=0 \tag{2.32a}$$

$$\chi = \hat{\Gamma} t^{-1}|\ln t|^{1/3}, \quad t>0, \quad H=0 \tag{2.32b}$$

$$\chi = \hat{\Gamma}'(-t)^{-1}|\ln|t||^{1/3}, \quad t=0, \quad H=0 \tag{2.32c}$$

$$C_H = \hat{A}|\ln|t||^{1/3}, \quad t>0, \quad H=0 \tag{2.32d}$$

$$C_H = \hat{A}'|\ln|t||^{1/3}, \quad t<0, \quad H=0 \tag{2.32e}$$

$$H = \hat{D}M^3|\ln|M||^{-1}, \quad t=0 \tag{2.32f}$$

instead of (2.6)–(2.11). With these definitions of amplitudes, one can again find four universal relations among the six amplitudes, and define $\hat{A}/\hat{A}'$, $\hat{\Gamma}/\hat{\Gamma}'$, $R_\chi$, and $R_C$ in terms of $\hat{A}$, $\hat{A}'$, etc.

## III. $\epsilon$ EXPANSIONS AND RENORMALIZATION-GROUP RESULTS

Some insight into the origins of the universality of the ratios (2.23)–(2.26) can be obtained from a derivation of the equation of state using the recursion relations near and at four dimensions.[17] Since this derivation is rather technical, we summarize it in Appendix A. A more direct calculation is to use the Feynman-graph expansion of the universal equation of state $\tilde{h}(\tilde{x})$, as derived by Brézin, Wallace, and Wilson[11] and Avdeeva and Migdal.[12] From this equation, using Eqs. (2.21) and (2.22), an explicit (although somewhat lengthy) calculation yields

$$\tilde{A} = \frac{n\epsilon}{4(n+8)}\left[1+\epsilon\left(1+\frac{6\ln 2 - \frac{9}{2}\ln 3}{n+8} - \frac{30}{(n+8)^2}\right)\right] + O(\epsilon) \tag{3.1}$$

and[18]

$$\Gamma^{-1}=1+\frac{3\epsilon}{2(n+8)}\ln\tfrac{4}{27}+\left(\frac{\epsilon}{2(n+8)}\right)^2\left(\tfrac{9}{2}\ln^2\tfrac{4}{27}+\frac{188+38n-n^2}{n+8}\ln\tfrac{4}{27}-2(n-1)S\right)$$
$$\simeq 1-\frac{2.8643\epsilon}{n+8}+\frac{\epsilon^2}{(n+8)^2}\left(4.1022-0.4774\,\frac{188+38n-n^2}{n+8}+4.9249(n-1)\right), \tag{3.2}$$

where

$$S=I_1(\tfrac{1}{4})-6I_1(\tfrac{3}{4})-3I_2(\tfrac{3}{4})+2I_3(\tfrac{3}{4})-\tfrac{1}{4}\ln^2 3, \tag{3.3}$$

with $I_1$, $I_2$, and $I_3$ defined in Ref. 11.

Combining (2.26), (3.1), and (3.3) we immediately find

$$R_C=\frac{n\epsilon 2^{-2\beta-1}}{n+8}\left[1+\epsilon\left(1-\frac{30}{(n+8)^2}\right)\right]+O(\epsilon), \tag{3.4}$$

where

$$2\beta=1-\frac{3\epsilon}{n+8}+O(\epsilon^2) \tag{3.5}$$

and $2^{-2\beta}$ means

$$2^{-2\beta}=\tfrac{1}{2}\left(1+\frac{3\epsilon}{n+8}\ln 2+O(\epsilon^2)\right). \tag{3.6}$$

Similar powers of 2 appear in the $\epsilon$ expansions of $A/A'$ and $\Gamma/\Gamma'$,[19] originating from the different basic temperature scales above and below $T_c$. They would disappear from combinations like $A'B^{-2}\Gamma'$, when *all* the amplitudes are for $T<T_c$.

Similarly, (2.25) and (3.2) immediately yield $R_\chi=\tilde{\Gamma}$. For completeness we also list the $\epsilon$ expansions of $A/A'$ and $\Gamma/\Gamma'$ first derived by Brézin *et al.*,[19]

$$A/A'=\tilde{A}/\tilde{A}'=2^{\alpha-2}(1+\epsilon)n+O(\epsilon^2), \tag{3.7}$$

$$\Gamma/\Gamma'=\tilde{\Gamma}/\tilde{\Gamma}'=2^{\gamma-1}\gamma/\beta+O(\epsilon^3)\quad\text{for } n=1. \tag{3.8}$$

The $\epsilon$ expansions of $\alpha$, $\beta$, $\gamma$, etc., are given in Ref. 9. Note that $\tilde{\Gamma}'=\infty$ for $n>1$.

There are many ways to extrapolate the expansions (3.2) and (3.4) to $\epsilon=1$. Clearly, using the expansion (3.4) for $\tilde{\Gamma}^{-1}$ with $\epsilon=1$ and then inverting the result, or inverting the expansion and then substituting $\epsilon=1$ yield different numerical results. For example, for $n=1$,[18]

$$\tilde{\Gamma}^{-1}=1-0.3183\epsilon-0.0967\epsilon^2\xrightarrow[\epsilon=1]{}0.5850=\frac{1}{1.709}, \tag{3.9}$$

whereas

$$\tilde{\Gamma}=1+0.3183\epsilon+0.1980\epsilon^2\xrightarrow[\epsilon=1]{}1.516. \tag{3.10}$$

A third way is to expand $\ln\tilde{\Gamma}$, and then put $\epsilon=1$. At least this way one will get consistent results when using $\tilde{\Gamma}$ or $\tilde{\Gamma}^{-1}$. This yields

$$\tilde{\Gamma}=\exp(0.3183\epsilon+0.1473\epsilon^2)\xrightarrow[\epsilon=1]{}1.593. \tag{3.11}$$

We shall use the spread in values thus obtained as a rough estimate of the errors of the extrapolation procedure. For $n=1$, $\epsilon=1$ we have

$$R_\chi=\tilde{\Gamma}=1.61\pm0.10,\quad n=1,\ d=3. \tag{3.12}$$

A similar study of the case $n=3$ yields

$$R_\chi=\tilde{\Gamma}=1.33\pm0.01,\quad n=3,\ d=3. \tag{3.13}$$

The situation with regard to $R_C$ is similar: One can expand $R_C$, $R_C^{-1}$, or $\ln(R_C/\epsilon)$, one can exponentiate $2^{-2\beta}$ or expand it, one can use the known value of $\beta$, from series, or the one resulting from the first-order $\epsilon$ expansion (3.5), etc. Since the coefficient of $\epsilon$ in Eq. (3.4) is close to $+1$, the result from calculating $R_C^{-1}$ is probably less reliable. The different methods of extrapolation of (3.4) [with $2\beta$ in the exponent taken from (3.5)] yield for $n=1$, $d=3$,

$$R_C=\frac{\epsilon}{36}(1+0.861\epsilon)\to0.052,$$
$$R_C=\frac{\epsilon}{36(1-0.861\epsilon)}\to0.2, \tag{3.14}$$
$$R_C=\frac{\epsilon}{36}\exp(0.861\epsilon)\to0.066,$$

and for $n=3$, $d=3$,

$$R_C=\frac{3\epsilon}{44}(1+0.941\epsilon)\to0.132,$$
$$R_C=\frac{3\epsilon}{44(1-0.941\epsilon)}\to1.16, \tag{3.15}$$
$$R_C=\frac{3\epsilon}{44}\exp(0.941\epsilon)\to0.175.$$

Note that $R_C=0$ for $n=0$, and

$$\frac{R_C}{n}=\frac{\epsilon}{(2-\epsilon)^2}=\frac{\epsilon}{4}(1+\epsilon)+O(\epsilon^2) \tag{3.16}$$

for the spherical model ($n=\infty$) (see Appendix B). Thus $R_C$ is a smooth function of $n$ near $d=4$. From a study of series expansions in Sec. IV, we shall see that $R_C$ is also monotonic for $d=3$, and that it varies from 0 to $\infty$ as $n$ goes from 0 to $\infty$. This is in contrast to the quantity[8] $R=\alpha^{-1}R_C$, which is directly proportional to the specific heat [see Eqs. (2.28) and (2.31b), but diverges when $\alpha=0$, and changes sign at that point. Thus the theoretical uncertainties are much smaller for quantitative estimates of $R_C$, than they are for $\alpha^{-1}R_C$.

Similar to the above analysis, one can extrapolate the expressions (3.7) and (3.8) for $A/A'$ and

$\Gamma/\Gamma'$ to $\epsilon=1$ in various ways. Since the coefficients in the $\epsilon$ expansion (3.8) of $\Gamma/\Gamma'$ are not very large, all the extrapolations yield values close to[19]

$$\Gamma/\Gamma' \approx 4.8, \quad n=1, \ d=3. \tag{3.17}$$

On the other hand, the coefficient of $\epsilon$ in (3.7) is equal to $+1$, so that replacement of $(1+\epsilon)$ by $(1-\epsilon)^{-1}$ leads to divergence at $\epsilon=1$. Extrapolation of (3.7) yields[19]

$$A/A' \approx \begin{cases} 0.55, & n=1, \ d=3 \\ 1.36, & n=3, \ d=3 \end{cases} \tag{3.18}$$

while extrapolation of $\ln(A/A')$ will increase these results by a factor 1.36.

As noted by Brézin *et al.*,[19] the results (3.18) are in surprisingly good agreement with those obtained in specific-heat experiments.[5,7] Nevertheless, the ambiguities in the $\epsilon$ expansion for $A/A'$ noted above would suggest that one should regard the agreement with considerable caution. It would therefore be highly desirable to have the next order in $\epsilon$, in order to use Padé approximants, which might give greater consistency between the various extrapolations. Similar comments apply to the results (3.14) and (3.15): The first and third extrapolations are in reasonable agreement with the series values discussed in Sec. IV, which leads us to conclude that the second extrapolation should not be used. The values we choose as best extrapolations of the $\epsilon$ expansion at $\epsilon=1$ are summarized in Table I.

Before concluding this section, we give some results for dipolar systems. In analogy to the above discussion, one can use the dipolar equation of state for $n=d=4-\epsilon$ (Ref. 20) to obtain all the amplitude ratios. Since $\Gamma'=\infty$, $\Gamma/\Gamma'$ is meaningless. Reference 20 already includes the results

$$A/A' = \tfrac{6}{5} + O(\epsilon) \quad \text{(dipolar)} \tag{3.19}$$

and

$$R_\chi = \tilde{\Gamma} = 1 + \tfrac{1}{4}\epsilon + O(\epsilon^2) \quad \text{(dipolar)}\ . \tag{3.20}$$

A similar calculation yields

$$R_C = \tfrac{3}{34}\epsilon + O(\epsilon^2) \quad \text{(dipolar)}\ . \tag{3.21}$$

Since these results are available only to the lowest nontrivial order, it is difficult to compare them with real systems at $d=3$. However, one can compare these expansions to their short-range counterparts, Eqs. (3.7), (3.2), and (3.4), which yield (for $n=d=4-\epsilon$)

$$A/A' = 1 + O(\epsilon)\ ,$$

$$R_\chi = 1 + 0.239\epsilon + O(\epsilon^2)\ ,$$

$$R_C = \tfrac{1}{12}\epsilon \quad \text{(short range, } n=d)\ . \tag{3.22}$$

At $\epsilon=1$, the two sets of values do not differ significantly from each other. (The apparent difference in $A/A'$ may be an artifact due to the vanishing of $\alpha$ to lowest order in $\epsilon$, for the short-range, $n=d$ case.)

The calculation for the dipolar Ising ($n=1$) case is different, since the critical behavior is now described by Eq. (2.32), i.e., by mean-field expressions with logarithmic corrections. The easiest way to treat this case is to use the recursion relations, as described in Appendix A. Most of the amplitude ratios for this case were obtained by Brézin,[21] with the results

$$\hat{A}/\hat{A}' = \tfrac{1}{4}, \quad \hat{\Gamma}/\hat{\Gamma}' = 2\ , \tag{3.23}$$

and[14]

$$Q_1 = 3\hat{\Gamma}_c(\hat{B}^2\hat{\Gamma})^{-1/3} = (\tfrac{2}{3})^{1/3}\ , \tag{3.24}$$

where $\hat{\Gamma}_c$ is the amplitude of the susceptibility along the critical isotherm,

$$\chi = \hat{\Gamma}_c H^{-2/3}|\ln H|^{1/3}, \quad t=0\ . \tag{3.25}$$

Combining (2.23) and (3.25), we see that $\hat{\Gamma}_c = (81\hat{D})^{-1/3}$, and hence

$$R_\chi = \hat{D}\hat{\Gamma}\hat{B}^2 = \tfrac{1}{3}Q_1^{-3} = \tfrac{1}{2}\ . \tag{3.26}$$

TABLE I. Summary of $d=3$ values of thermodynamic amplitude ratios for various models.

| | $n=0$ | $n=1$ | | $n=3$ | | $n=1$ | Mean |
|---|---|---|---|---|---|---|---|
| | $\epsilon$ expansion | Series[a] | $\epsilon$ expansion | Series[a] | $\epsilon$ expansion | Dipolar | field theory |
| $A/A'$ | 0 | 0.51 | 0.55 | 1.52 | 1.36 | $\frac{1}{4}$ | 0[b] |
| $\Gamma/\Gamma'$ | | 5.07 | 4.80 | ... | ... | 2 | 2 |
| $R_C$ | 0 | 0.059 | 0.066 | 0.165 | 0.17 | $\frac{1}{6}$ | 0[b] |
| $R_\chi$ | 1.9 | 1.75 | 1.6 | 1.23 | 1.33 | $\frac{1}{2}$ | 1 |

[a] See Table III.
[b] See Eq. (3.28), and preceding discussion.

A similar calculation, along the lines described in Appendix A, yields[22]

$$R_C = \hat{A}\hat{B}^{-2}\hat{\Gamma} = \tfrac{1}{6} \quad . \tag{3.27}$$

As noted by Brézin,[21] the result (3.26) is not given by the limit $\epsilon \to 0$ of (3.2), although (3.23) and (3.27) are given by the $\epsilon \to 0$ limit of (3.7) and (3.4). This is simply due to the logarithmic factors in Eq. (2.32), which affect the scales of $\ln M$ and $\ln H$ along the critical isotherm.

In mean-field theory, the amplitude $A$ vanishes, but the jump in specific heat $\Delta C$ is related to the other critical amplitudes by the universal ratio.

$$\Delta C\,\Gamma B^{-2} = \tfrac{1}{2} \ , \tag{3.28}$$

which is analogous to $A'\Gamma B^{-2} = R_C A'/A$. Note, however, that the value of (3.28) does not agree with the corresponding expression $\hat{A}'\hat{\Gamma}\hat{B}^{-2} = \frac{2}{3}$ of the three-dimensional dipolar Ising model.

## IV. SERIES EXPANSIONS AND REAL SYSTEMS

### A. Ising model

In two dimensions the amplitudes $B$, $\Gamma$, and $\Gamma'$ have been calculated exactly,[23] while $A$, $A'$, and $D$ have been obtained by series.[24] The results for the amplitude ratios are shown in Table II.

In three dimensions, the amplitudes $\Gamma$, $\Gamma'$, $B$, $D$, and $A$ have been calculated for a number of lattices using series-expansion techniques.[25] The ensuing values of the universal ratios have roughly a 5% spread between the different lattices. We show a typical set of values in Table III. For the amplitude $A'$ the low-temperature series seem quite unreliable,[5] and we quote instead the value of $A/A'$ obtained in Ref. 5 by integration of the equation of state of Gaunt and Domb.[26]

### B. Heisenberg model

For the case $n=3$, there exist only high-temperature series, and we use the values of $B$, $\Gamma$, and $D$ obtained for the $S=\infty$, fcc lattice by Miloševič and Stanley[27] (see Table III). There also exists a Monte Carlo calculation for the classical sc Heisenberg model by Binder and Müller-Krumbhaar,[28] with parameters values $\gamma = 1.36$, $\delta = 5.3$, $B = 1.03$, $\Gamma = 0.32$, $D = 4.74$, which yield $R_\chi = 1.73$. This number is not particularly close to the series and $\epsilon$-expansion values quoted in Table III. The amplitude $A$ was given by Stauffer *et al.*[29]

As was mentioned already in discussing the Ising case, series determinations of specific-heat amplitudes seem rather unreliable, and it is preferable to obtain both $A$ and $A'$ by integrating the equation of state.[5] Such a procedure was followed by Krasnow and Stanley[30] in the Heisenberg model, but the ensuing value of $A$ disagrees strongly with the value quoted in Ref. 29, and leads to improbable values of $R_C$. We have therefore discarded this evaluation, assuming that some error had been made, as was already found[5] in the Ising case. Similarly, the $A'$ found in Ref. 30 cannot be relied on, even though the $A/A'$ has the "reasonable" value 1.46. It would be interesting to re-evaluate $A$ and $A'$ from the Heisenberg equation of state of Miloševič and Stanley, since we have no reason to doubt its validity, but we have not carried out the necessary integration.

As noted in Sec. III, the $\epsilon$ expansions for $\tilde{A}$ and $A/A'$ also seem particularly ill behaved. Therefore, we believe that the most reliable way to estimate $A/A'$ comes from the observation[5,7] that for small $\epsilon$ one can write

$$A/A' \simeq 1 - \mathcal{P}\,\alpha \ , \tag{4.1}$$

TABLE II. Universal amplitude ratios for the two-dimensional Ising model (Refs. 23 and 24).

| |
|---|
| $A/A' = 1$ |
| $\Gamma/\Gamma' = 37.693\,651\,95$ |
| $R_\chi = 6.780$ |
| $R_C = 0.318\,57$ [a] |

[a] Since the specific heat diverges logarithmically, $(t^{-\alpha}-1)/\alpha$ is replaced by $-\ln t$. The definition of $R_C$, Eq. (2.26), remains unchanged.

TABLE III. Series values for amplitudes and universal ratios for typical three-dimensional models.

| | Ising $n=1$, bcc | Heisenberg $n=3$, $s=\infty$, fcc |
|---|---|---|
| $A$ | 0.0138 [a] | 0.88 [b] |
| $A'$ | 0.027 [a] | 0.58 [c] |
| $B$ | 1.5059 [d] | 1.22 [e] |
| $\Gamma$ | 0.985 [d] | 0.279 [e] |
| $\Gamma'$ | 0.194 [d] | $\infty$ |
| $D$ | 0.345 [d] | 2.1284 [e] |
| $\alpha$ | $\frac{1}{8}$ [d] | $-0.13$ [e] |
| $\beta$ | $\frac{5}{16}$ [d] | 0.38 [e] |
| $A/A'$ | 0.51 | 1.52 [f] |
| $\Gamma/\Gamma'$ | 5.07 | ... |
| $R_C$ | 0.059 | 0.165 |
| $R_\chi$ | 1.75 | 1.23 |

[a] From integrating the equation of state; see Ref. 5.
[b] Reference 29.
[c] Obtained from $A/A'$ and from $A$.
[d] References 25 and 26.
[e] Reference 27.
[f] From Eq. (4.1).

with $\mathcal{P} \approx 4$, independent of $\alpha$. We have therefore used this method of obtaining the "series" value of $A/A'$ quoted in Table I for $n=3$ (with $\alpha$ determined from series), but clearly a more direct evaluation would be highly desirable. Our three-dimensional estimates of the ratios coming from $\epsilon$ expansions and series are summarized in Table I for various models.

### C. Real systems

In this section we first wish to summarize briefly the information on universal amplitudes which was obtained in the analysis of four systems (Xe, $CO_2$, Ni, and EuO) in Ref. 5. It must be emphasized that this analysis was by no means complete, since results from various experiments were combined, and no detailed consistency checks were made. Thus the data presented here are primarily illustrative of the type of analysis which is feasible, rather than an attempt at accurate determinations of the universal amplitude ratios of real systems.

The data are summarized in Table IV, which contains the thermodynamic amplitude ratios obtained for various empirical equations of state. The parameters of these equations were determined by fits to experimental exponents and amplitudes. As explained in Ref. 5, these "experimental" quantities are not necessarily the most accurate representation of the raw data, since various constraints had been placed on the fits, such as the scaling relations (2.12), or the assumption that there are no singular corrections to scaling. The linear-model (LM) and Missoni-Levelt-Sengers-Green (MLSG) equations contain two scales, determined by fits to $B$ and $\Gamma$, and three universal parameters. These are two independent exponents (usually $\beta$ and $\delta$), and one extra quantity, denoted $b$ or $E_2$, which is fixed by an over-all fit to the equation of state. These five parameters entirely determine the equation of state and yield the amplitude ratios quoted in Table IV. As was noted in Ref. 5, the values obtained for $A$ and $A'$ in the linear-model analysis of EuO and Ni are quite far from the experimental values. The "modified MLSG" equation has an extra universal parameter ($e_3$), which was determined in Ref. 5 by a fit to the experimental $A/A'$. The independent exponents used in that analysis were $\beta$ and $\alpha$, with $\gamma$ and $\delta$ determined by scaling. The precise values of $\beta$ and $\alpha$ chosen were such that the amplitude $A$ itself fit experiment. Such choices were possible within the uncertainties of the data, but this would probably no longer be the case with better data, and a larger number of universal parameters would have to be introduced into the empirical equation of state, in order to obtain an adequate fit.

From the numerical results for the critical amplitude ratios in Table IV, we see that there is some consistency between the various empirical equations for a given substance, and also some consistency between fluids on the one hand and magnets on the other. The ratios $R_\chi$ and $R_C$ roughly follow the theoretical trends in Table I, in going from $n=1$ to $n=3$. The ratio $A/A'$ seems quite well behaved, as was discussed in detail in

TABLE IV. Real systems (Ref. 5).

| | $CO_2$ | | | Xe | | | Ni | | EuO | |
|---|---|---|---|---|---|---|---|---|---|---|
| | LM | MLSG | Modified MLSG | LM | MLSG | Modified MLSG | LM | Modified MLSG | LM | Modified MLSG |
| $\alpha$ | 0.10 | 0.10 | 0.10 [a] | 0.09 | 0.07 | 0.11 [a] | −0.11 | −0.09 [a] | −0.06 | −0.04 [a] |
| $\beta$ | 0.35 [a] | 0.35 [a] | 0.35 [a] | 0.35 [a] | 0.35 [a] | 0.36 [a] | 0.38 [a] | 0.38 [a] | 0.39 [a] | 0.37 [a] |
| $\gamma$ | 1.20 | 1.20 | 1.21 | 1.21 | 1.24 | 1.18 | 1.35 [a] | 1.33 | 1.29 [a] | 1.31 |
| $\delta$ | 4.44 [a] | 4.44 [a] | 4.48 | 4.46 [a] | 4.53 [a] | 4.32 | 4.58 | 4.52 | 4.35 | 4.56 |
| $A$ | 2.2 | 2.5 | 3.2 [a] | 2.1 | 2.9 | 1.6 [a] | 0.17 | 0.23 [a] | 0.25 | 0.47 [a] |
| $A'$ | 5.1 | 5.5 | 6.6 [a] | 3.9 | 4.4 | 3.5 [a] | 0.081 | 0.17 [a] | 0.16 | 0.39 [a] |
| $B$ | 2.0 [a] | 2.0 [a] | 2.0 [a] | 1.8 [a] | 1.8 [a] | 1.8 [a,b] | 1.5 [a] | 1.4 [a] | 1.3 [a] | 1.3 [a] |
| $\Gamma$ | 0.065 [a] | 0.065 [a] | 0.061 [a] | 0.074 [a] | 0.065 [a] | 0.12 [a,b] | 1.5 [a] | 1.3 [a] | 0.55 [a] | 0.37 |
| $\Gamma'$ | 0.014 | 0.015 | 0.015 | 0.018 | 0.016 | 0.028 | 0.38 | 1.1 | 0.15 | 0.14 |
| $D$ | 2.4 | 2.3 | 2.3 | 2.7 | 3.0 | 2.7 | 0.29 | 0.29 | 1.3 | 1.8 |
| $R_\chi$ | 1.6 | 1.6 | 1.5 | 1.6 | 1.6 | 2.1 | 1.7 | 1.4 | 1.6 | 1.5 |
| $R_C$ | 0.037 | 0.041 | 0.050 | 0.048 | 0.058 | 0.061 | 0.11 | 0.16 | 0.18 | 0.11 |
| $\alpha^{-1}R_C$ | 0.35 | 0.39 | 0.53 | 0.54 | 0.89 | 0.56 | −1.0 | −1.7 | −3.0 | −2.5 |
| $A/A'$ | 0.43 | 0.45 | 0.54 [a] | 0.54 | 0.65 | 0.44 [a] | 2.0 | 1.4 [a] | 1.6 | 1.2 [a] |
| $\Gamma/\Gamma'$ | 4.6 | 4.2 | 4.0 | 4.2 | 4.1 | 4.3 | 3.9 | 1.3 | 3.7 | 2.7 |

[a] Experimental input to the equation of state; see Ref. 5.

[b] This number was incorrect in Table V of Ref. 5.

Ref. 5, and $\Gamma/\Gamma'$ is reasonably consistent for fluids, but quite erratic in the magnets. As mentioned in Ref. 5, its physical meaning is not clear in that case, since the measured values are dominated by corrections to the Heisenberg Hamiltonian. We should also mention that EuO is expected to show more "dipolar" behavior than Ni,[31] but we do not feel that there is accurate enough experimental or theoretical information at this stage to make meaningful comparisons of the amplitude ratios.

Another real system which has recently become of great interest, is the dipolar Ising ferromagnet $LiTbF_4$, for which careful measurements of the specific heat[32] confirm the form (2.32), with $\hat{A}/R \approx 0.4394$. Data for the susceptibility above $T_c$ were fit to[33]

$$\chi/\chi_0 \approx 1.25\, t^{-1.13}, \quad t>0,\ H=0$$

while data for the magnetization were fit to[34]

$$M/M_0 \approx 2.135(-t)^{0.45}, \quad t<0,\ H=0 \ .$$

Forcing these results into the forms given in (2.32), we find that roughly $\hat{B}/M_0 \approx 2.92$, $\Gamma/\chi_0 \approx 2.8$, so that finally $R_c \approx 0.14$, in reasonable agreement with the exact prediction (3.28). It should be remembered, however, that these fits are very crude, and a direct analysis of the original data, including corrections to the leading singular terms,[22] would provide a better test of the theory.

In concluding this brief summary of experimental data, our over-all impression is that the question of universal amplitude ratios in real systems is still at a very crude stage, and much more careful work needs to be done before our knowledge of the ratios approaches that of the critical exponents.

In Table I we also included the values of the amplitude ratios for $n=0$. It would be interesting to relate these to experiments (and theories) on polymer solutions.[35]

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## APPENDIX A: RENORMALIZATION GROUP

In this appendix, we use the renormalization-group recursion relation results of Nelson and Rudnick[17] to derive the universal amplitude ratios. They start with the reduced Ginzburg-Landau-Wilson Hamiltonian

$$\overline{\mathcal{H}} = -\int d^d x \{ \tfrac{1}{2}[r\vec{S}(\vec{x})^2 + (\vec{\nabla}\vec{S})^2] + u\vec{S}(\vec{x})^4 - HS_n(\vec{x})\} \ . \qquad \text{(A1)}$$

They then solve the recursion relations for $r$, $u$, $H$, and the magnetization $M = \langle S_n \rangle$, and stop iterating after $l^*$ iterations, such that the longitudinal susceptibility $\chi(l^*)$ is equal to unity. At this point, they calculate the various thermodynamic functions using perturbation theory. The magnetization, field, and longitudinal susceptibility are given by

$$M = e^{-(1-\epsilon/2)l^*} M(l^*) \ , \quad H = e^{-(3-\epsilon/2)l^*} H(l^*) \ ,$$
$$\chi = e^{2l^*}\chi(l^*) = e^{2l^*} \ , \qquad \text{(A2)}$$

while the perturbation expansion yields

$$H(l^*)/M(l^*) = t(l^*) + 4u(l^*)M(l^*)^2 + O(u(l^*)), \qquad \text{(A3)}$$

$$1 = \chi(l^*)^{-1} = t(l^*) + 12u(l^*)M(l^*)^2 + O(u(l^*)), \qquad \text{(A4)}$$

with (to order $\epsilon$)

$$u(l) = ue^{\epsilon l}/Q(l) \ ,$$
$$t(l) = te^{2l}Q(l)^{-(n+2)/(n+8)} \ ,$$
$$Q(l) = 1 + 4K_4(n+8)u(e^{\epsilon l} - 1)/\epsilon \ . \qquad \text{(A5)}$$

Determining $l^*$ from (A4), and substituting (A5), one can now obtain all the amplitudes in (2.6)–(2.11) to order $\epsilon$, and combine them to yield the ratios (2.23)–(2.26). It turns out that the non-universal parameter $u$ (and the momentum cutoff) drop out, to leave universal results for these ratios, in agreement with the ones quoted in Sec. III.

At $d=4\,(\epsilon=0)$, $Q(l) \approx 4K_4(n+8)ul$. Solving (A4) for $l^*$ yields

$$l^* \approx |\ln|M|| \quad \text{at } t=0,$$

or

$$l^* \approx \tfrac{1}{2}|\ln|t|| \quad \text{at } t<0,\ H=0.$$

Substituting these back into (A3) finally leads to the expressions (2.32). Combining the appropriate amplitudes yields (3.23), (3.27), and (3.28). To obtain (3.28) one also needs the free energy, which was found by Nelson and Rudnick[17] to be

$$F = -[nt^2/16u(4-n)](Q^{(4-n)/(n+8)} - 1) + \min_M(\tfrac{1}{2}tQ^{-(n+2)/(n+8)}M^2 + uQ^{-1}M^4 - HM) \ . \qquad \text{(A6)}$$

The Hamiltonian of the three-dimensional dipolar Ising model may be written,[15,16]

$$\bar{\mathcal{H}} = -\tfrac{1}{2}\int_{\vec{q}} [r+g^2+g(q_z/q)^2]\sigma_{\vec{q}}\sigma_{-\vec{q}} - u\int_{\vec{q}}\int_{\vec{q}'}\int_{\vec{q}''}\sigma_{\vec{q}}\sigma_{\vec{q}'}\sigma_{\vec{q}''}\sigma_{-\vec{q}-\vec{q}'-\vec{q}''} + H\sigma_0\ , \tag{A7}$$

where $\int_{\vec{q}} = (2\pi)^{-3}\int d^3q$, $|\vec{q}|<1$. The leading terms in the recursion relations for this case are completely analogous to their $d=4$ short-range counterparts, provided $u$ is replaced by $ug^{-1/2}$ and $\epsilon$ is replaced by $\epsilon-1$.[16] These terms are all we need to obtain the leading singularities, Eqs. (2.32a)–(2.32f). Thus, the final results (3.23), (3.27), and (3.28) are the same as those for the $d=4$ short-range case.

## APPENDIX B: SPHERICAL MODEL

There are several reasons why it is instructive to consider the exactly soluble spherical model. The main one is that the free energy per spin component of an $n$-component spin model approaches that of the spherical model in the limit $n\to\infty$.[36] One can thus calculate exactly the various universal quantities in this limit for any dimensionality. A comparison with the $\epsilon$ expansion is then helpful in obtaining a feeling about the accuracy of this expansion at $d=3$.

The universal equation of state in the spherical model is[37]

$$\tilde{h}(\tilde{x}) = (1+\tilde{x})^\gamma\ , \tag{B1}$$

with

$$\gamma = (1-\epsilon/2)^{-1},\quad \beta=\tfrac{1}{2}\ , \tag{B2}$$

and all other exponents obtained by scaling relations. In particular,

$$\alpha = -\frac{\epsilon}{2-\epsilon} < 0\ . \tag{B3}$$

Therefore, (2.19) must be modified to read[5]

$$\begin{aligned}\tilde{A} &= \alpha\beta(1-\alpha)(2-\alpha)\int_0^\infty dy\, y^{\alpha-3}[(1+y)^\gamma - 1 - \gamma y - \tfrac{1}{2}\gamma(\gamma-1)y^2]\ ,\\ \tilde{A}' &= -\alpha\beta(1-\alpha)(2-\alpha)\left(\frac{1}{2-\alpha} - \frac{\gamma}{1-\alpha} - \frac{\gamma(\gamma-1)}{2\alpha} + \int_0^{-1} dy\,|y|^{\alpha-3}[(1+y)^\gamma - 1 - \gamma y - \tfrac{1}{2}\gamma(\gamma-1)y^2]\right).\end{aligned} \tag{B4}$$

For $3<d<4$, these integrals yield

$$\tilde{A} = \epsilon/(2-\epsilon)^2,\quad \tilde{A}'=0\ . \tag{B5}$$

Equation (B1) directly yields

$$\tilde{B} = \tilde{D} = \tilde{\Gamma} = 1\ , \tag{B6}$$

and hence the result (3.16) for $R_C/n$. $\tilde{A}$ gives the specific heat per component,[36] and

$$R_\chi = 1\ . \tag{B7}$$

The result (B5) implies that $A/A'=\infty$ for $3<d<4$. Since $\alpha$ equals $-1$ at $d=3$, the calculation (B4) must be modified in that case. Equation (B1) becomes analytic in $\tilde{x}$ ($\gamma=2$), and the "singular" part in the free energy mixes with the "analytic" part. This leads to an arbitrariness in the definitions of $\tilde{A}$ and $\tilde{A}'$. The arbitrariness may be removed, however, if one considers the $n$-vector model, for very large (but *finite*) $n$, instead of the ($n=\infty$) spherical model. This was recently done by Abe and Hikami.[38] To leading order in $1/n$ they found, for $3<d<4$,

$$\frac{A}{A'} = \frac{n2^{d(3-d)/(d-2)}\Gamma(d/2)\Gamma(2-d/2)}{\Gamma((4-d)/(d-2))\Gamma((2d-6)/(d-2))} \times\left(\frac{\Gamma(\frac{3}{2})\Gamma(d/2)}{\Gamma((d-1)/2)}\right)^{d/(d-2)}\ , \tag{B8}$$

which agrees with (3.7) for $n\to\infty$, $\epsilon=4-d\to0$, and with $A/A'=\infty$ for $n=\infty$. At $d=3$, however, they found

$$A/A' = (\pi^2/4) - 1\ , \tag{B9}$$

which is not proportional to $n$. Thus, the limit $n\to\infty$ of $A/A'$ has a discontinuity at $d=3$, which makes comparison with the $\epsilon$ expansion at $\epsilon=1$ meaningless.

Returning to the result (3.16), it is clear that truncation of the $\epsilon$ expansion after one term, and using it for $\epsilon=1$, gives an error of 100%. This is mainly due to the large coefficient of $\epsilon$. This is another indication of the fact noted in Sec. III, that the $\epsilon$ expansion of $R_C$ may not be reliable at $\epsilon=1$.

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