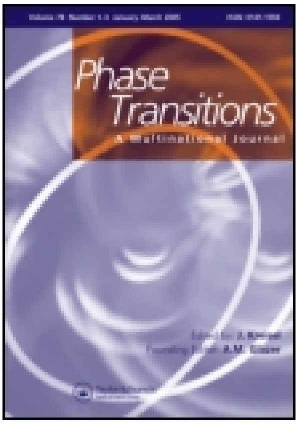

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Three-dimensional (3D) Ising universality in magnets and critical indices at fluid-fluid phase transition

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Three-dimensional (3D) Ising universality in magnets and critical indices at fluid–fluid phase transition

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Experimental data for critical exponents in some magnetic materials are compared with recent theoretical results on the three-dimensional (3D) Ising model, as derived by one of us (ZDZ) based on two conjectures [Z.D. Zhang, *Conjectures on the exact solution of three-dimensional (3D) simple orthorhombic Ising lattices*, Phil. Mag. 87 (2007), pp. 5309–5419]. It is found that critical exponents in some bulk magnetic materials indeed form a 3D Ising universality. Our attention is then focused on the critical indices at fluid–fluid phase transition. We suggest to use Zhang's exponent $\beta = 3/8$ to fit the experimental data over the wider asymptotic region near the critical point of a fluid–fluid phase transition. The 3D Ising universality should exist for critical indices in a certain class of magnets and at fluid–fluid phase transition.

Keywords: magnetically ordered materials; fluid–fluid; phase transitions; order–disorder effects

PACS: 75.40.-s; 75.40.Cx; 75.50.Cc

1. Introduction

Recently, the putative exact solution of the three-dimensional (3D) Ising model was obtained by one of us (ZDZ) introducing two conjectures and the critical exponents were derived to be $\alpha = 0$, $\beta = 3/8$, $\gamma = 5/4$, $\delta = 13/3$, $\eta = 1/8$, and $\nu = 2/3$ [1]. In ref. [2], March and Zhang asserted that, near criticality, the critical exponents β, γ, and δ given experimentally by Ho and Litster [3,4] for $CrBr_3$ agree to excellent accuracy with the conjectured ones [1]. Comparing with the experimental data for Ni(1 1 1) films on a W(1 1 0) substrate [5,6], March and Zhang found in [7] that there is a crossover from three to two dimensions between Zhang and Yang's critical exponents β with decrease in the thickness t of Ni films. Phase transitions of the mixed spin-1/2 and spin-S ($S \geq 1/2$) Ising model on a 3D decorated lattice with a layered magnetic structure were investigated within the framework of a precise mapping relationship to the simple spin-1/2 Ising model on a tetragonal lattice by adopting Zhang's solution [8]. Very recently, Ławrynowicz et al. [9] reformulated the algebraic part of the theory in terms of the quaternionic sequence of Jordan algebras to

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look at some of the geometrical aspects of simple orthorhombic Ising lattices, and represented a mathematical outlook of fractals and chaos related to these 3D Ising lattices [10].

These motivate us, in this study, to first compare the recent theoretical 3D Ising results [1] with more experimental data in various magnetic materials. It is shown that a 3D Ising universality indeed exists in some magnetic materials, regardless of their detailed 3D structures. Our attention is then focused on the critical indices at fluid–fluid phase transition. After the analysis of the procedure for studying the critical behavior of fluid–fluid phase transition in literature, we suggest that one could attempt to apply a simple power law with Zhang's exponent $\beta = 3/8$ to fit the experimental data over a wider asymptotic region near the critical point of a fluid–fluid phase transition. It is expected that there will be a 3D Ising universality for critical indices in many magnets and at fluid–fluid phase transition.

2. 3D Ising universality in magnets

In magnetic materials, as the temperature or other variable approaches its critical point, physical quantities diverge to infinity or converge to zero [1]. For instance, in the region just below the critical point T_c, the spontaneous magnetization is described by a power law,

$$M \propto (T_c - T)^{\beta}. \tag{1}$$

The correlation length ξ approaches infinity as $T \to T_c$.

$$\xi(T) \sim (T - T_c)^{-\upsilon}. \tag{2}$$

The magnetic susceptibility can be described as

$$\chi \sim (T - T_c)^{-\gamma}. \tag{3}$$

Similarly, the magnetization at magnetic fields and the correlation function at the critical point T_c fall off as power laws with the critical exponents δ and η, respectively. However, the specific heat near the critical temperature may show either a power law with the critical exponent $\alpha < 0.2$ or a logarithmic singularity with $\alpha = 0$. It was understood that the behaviors of curves with a power law of $\alpha < 0.2$ are very close to the logarithms with $\alpha = 0$, so that usually, one cannot distinguish the difference between them [1].

The accuracies of experiments for the determinations of critical exponents could be reduced by many factors [1,11]. Vicentini-Missoni [11] pointed out, after a careful examination on data up to the early 1970s, that good data in the critical region are available only on few substances, i.e., the nickel data of Weiss and Forrer [12], and those of Kouvel and Comly [13], the gadolinium data of Graham Jr [14], and the $CrBr_3$ data of Ho and Litster [3,4,15]. The data (a) in table III of Vicentini-Missoni's review [11] were determined by least-squares fit of their experimental data [16–18], which agree well with the results in [1]. The critical indices $\gamma = 1.22 \pm 0.05$, $\delta = 4.4 \pm 0.2$, $\eta = 0.123$, and $\nu = 0.65 \pm 0.05$, given in Table 1. 3 of Kadanoff's chapters [38] for the real fluids (being the same 3D Ising universality) are also very close to Zhang's [1] solution. We shall collect more experimental data reported for the critical exponents in various magnetic materials in the last decades. Most of the data collected here are consistent with the recent 3D Ising results [1]. Furthermore, some early controversy in several materials would be resolved if one would accept the conjectured exponents.

Table 1. Experimental data reported for the critical exponents in various magnetic materials, which are compared with the theoretical values of the 3D Ising model [1].

	α	β	γ	δ	η	ν	References
3D Ising	0	3/8	5/4	13/3	1/8	2/3	[1]
$CrBr_3$		0.364 ± 0.005	1.21	4.32 ± 0.10			[3,4,15]
Gd		0.370 ± 0.010	1.25	4.39 ± 0.10			[8,14]
Gd		$0.37 - 0.385$					[19]
Gd			1.196				[20]
300 Å Gd(0 0 0 1)		0.375(6)					[21]
Melt-spun Gd		0.389 ± 0.017	1.300 ± 0.014	4.32 ± 0.02			[22]
Gd		0.376 ± 0.015					[23]
Gd		0.381	1.196	3.615			[24]
Ni		0.373 ± 0.016	1.28	4.44 ± 0.18			[11,13]
Ni		0.375 ± 0.013	1.31	4.48 ± 0.14			[11,12]
Ni		0.378	1.34	4.58			[24]
Ni			1.28 ± 0.03				[25]
Fe		0.389	1.33	4.35			[24]
Fe		0.37 ± 0.03	1.30 ± 0.06				[26]
Co		0.435	1.21	3.35			[24]
Co			1.21 ± 0.04				[27]
$Fe_{1.50}Mn_{1.50}Si$		0.383 ± 0.01	1.274 ± 0.06	4.45 ± 0.19			[28]
$Fe_{30}Tb_{70}$		0.370	1.402	4.779			[29]
$Fe_{33}Gd_{67}$		0.401	1.210	4.240			[29]
$Fe_{80}P_{13}C_7$ amorphous	-0.06 ± 0.09	0.38 ± 0.02	1.30 ± 0.05	4.47 ± 0.05			[30]
$Fe_{75.5}Cr_4B_{13}Si_{7.5}$ amorphous		0.366	1.286				[31]
$KMnF_3$			1.24 ± 0.5			0.62 ± 0.07	[32]
Rb_2ZnCl_4		0.35 ± 0.01	1.28 ± 0.09			0.66 ± 0.02	[33]
Rb_2ZnCl_4		0.350 ± 0.015	1.26 ± 0.04			0.683 ± 0.015	[34]
$Y_3Fe_5O_{12}$			1.228 ± 0.014				[35]
$NiCO_3$		0.365 ± 0.015					[35]
$LaMn_{0.95}Ti_{0.05}O_3$		0.378 ± 0.007	1.29 ± 0.02	4.19 ± 0.03			[36]
$LaMn_{0.9}Ti_{01}O_3$		0.375 ± 0.005	1.25 ± 0.02	4.11 ± 0.04			[36]
$LaMn_{0.85}Ti_{0.15}O_3$		0.376 ± 0.003	1.24 ± 0.01	4.16 ± 0.03			[36]
$LaMn_{0.8}Ti_{0.2}O_3$		0.359 ± 0.004	1.28 ± 0.01	4.21 ± 0.05			[36]
$La_{0.7}Ca_{0.3}MnO_3$		0.36 ± 0.01	1.2	4.263			[37]

The critical exponent $\beta = 0.37 - 0.385$ [19] was interpreted that Gd behaves as a Heisenberg system, while the critical exponent $\gamma = 1.196$ [20] for the paramagnetic susceptibility χ_0 indicates an Ising-like system. The possibility that this puzzle for bulk Gd may be caused by sample imperfections of the macroscopic specimens was excluded by measurement on ultraclean UHV-prepared Gd films [21]. Farle et al. [21] obtained $\beta = 0.375(6)$ for layer-by-layer grown 300 Å Gd(0 0 0 1) in agreement with bulk experiments ($\beta = 0.37 - 0.385$) [19]. Such discrepancy above for bulk Gd can be resolved in favor of a 3D Ising system with $\beta = 3/8$ and $\gamma = 5/4$ [1], although the results in [1] were obtained for the simple orthorhombic Ising lattices but Gd is in hexagonal close-packed (hcp) structure with space group $P6_3/mmc$. Melt-spun Gd is a structurally inhomogeneous system, which exhibits the exponents $\beta = 0.389 \pm 0.017$, $\gamma = 1.300 \pm 0.014$, and $\delta = 4.32 \pm 0.02$ [22]. Arnold and Pappas [23] claimed that the critical exponents for both surface, $\beta s = 0.83 \pm 0.04$, and bulk, $\beta_B = 0.376 \pm 0.015$, are consistent with the semi-infinite 3D Heisenberg model, but this bulk value is the same as the recent value $\beta = 3/8$ for the 3D Ising model [1].

The critical exponents β, γ, and δ of $Fe_{1.50}Mn_{1.50}Si$ alloy (with the $D0_3$ space group Fe_3Al-type structure) are 0.383 ± 0.01, 1.274 ± 0.06, and 4.45 ± 0.19, respectively [28], which are comparable with those for Fe, Co, and Ni [24] and close to the conjectured ones, $\beta = 3/8$, $\gamma = 5/4$, and $\delta = 13/3$ [1]. The wavelength and frequency dependence of neutrons magnetically scattered from iron was studied [26] and the critical exponents obtained are $\beta = 0.37 \pm 0.03$, $\gamma = 1.30 \pm 0.06$. Yamada et al. [30] reported the critical exponents as $\beta = 0.38 \pm 0.02$, $\gamma = 1.30 \pm 0.05$, $\delta = 4.47 \pm 0.05$, and $\alpha = -0.06 \pm 0.09$ for amorphous $Fe_{80}P_{13}C_7$. The critical exponents determined from magnetic susceptibility are $\gamma = 1.21 \pm 0.04$ for face-centered-cubic cobalt [27] and $\gamma = 1.28 \pm 0.03$ for nickel [25]. The critical exponents β, γ, and δ were determined by a modified Arrott plot, Kouvel–Fisher plot, Scaling plot, and ln (M) *versus* ln(H) plot [29]; β value (=0.370) of $Fe_{30}Tb_{70}$ and, γ and δ values ($\gamma = 1.210$, $\delta = 4.240$) of $Fe_{33}Gd_{67}$ obtained in [29] agree well with the conjectured values [1]. $Fe_{75.5}Cr_4B_{13}Si_{7.5}$ amorphous alloy has the critical exponents $\gamma = 1.286$ and $\beta = 0.366$ [31]. However, more attention is needed to pay on understanding why other amorphous alloys with different compositions have different critical exponents [31].

The critical exponents $\nu = 0.62 \pm 0.07$ and $\gamma = 1.24 \pm 0.5$ were obtained for $KMnF_3$ [32,39]. The critical exponents obtained for Rb_2ZnCl_4 in [33] are $\nu = 0.66 \pm 0.02$ and $\gamma = 1.28 \pm 0.09$, while those obtained in [34] are $\nu = 0.683 \pm 0.015$ and $\gamma = 1.26 \pm 0.04$. Although Rb_2ZnCl_4 was considered to belong to the 3D-XY universality class in [33], these critical exponents are closer to the recent 3D Ising values [1]. The critical exponent $\gamma = 1.228 \pm 0.014$ for YIG ($Y_3Fe_5O_{12}$) [35] is closer to the 3D Ising value $\gamma = 5/4$ [1] than to the mean field value $\gamma = 1$ or the Heisenberg value $\gamma = 4/3$. The critical exponent $\beta = 0.365 \pm 0.015$ was obtained for $NiCO_3$ [35].

The critical exponents, derived from the magnetic data of ferromagnetic $LaMn_{1-x}Ti_xO_3$ $(0.05 \le x \le 0.2)$ with rhombohedral structure ($R\bar{3}C$) [40], yield $0.359 \le \beta \le 0.378$, $1.24 \le \gamma \le 1.29$, and $4.11 \le \delta \le 4.21$ with a T_c of 95–173 K [36]. They suggested that β value is Heisenberg-like and γ value is 3D Ising model-like. But it is impossible that different critical exponents of a material behave as described, respectively, by two different models (similar situation can also be found in Ni films where a crossover from three to two dimensions between Zhang and Yang's critical exponents β with decreasing thickness can be only within the same model, i.e., 3D Ising model [7]). We argue here that $LaMn_{1-x}Ti_xO_3$ behaves as 3D Ising systems, according to the fact that both β and γ values are very close to the conjectured $\beta = 3/8$ and $\gamma = 5/4$ [1]. Similarly, the discrepancy for critical exponents $\beta = 0.36 \pm 0.01$ and $\gamma = 1.2$ of $La_{0.7}Ca_{0.3}MnO_3$ [37] can be resolved by the consideration that it belongs to 3D Ising category. Note that δ value determined using the Widom scaling equation $\delta = 1 + \gamma/\beta$ is 4.333 [37], the same as Zhang's theoretical value $\delta = 13/3$ [1], while the experimental value $\delta = 4.263$ determined from $\ln(M)$ *versus* $\ln(H)$ plot matches well with them [37].

It can be concluded that there is indeed a 3D Ising universality in some magnetic materials, independent of their detailed 3D structures, in good agreement with the prediction of [1] for the 3D Ising lattices.

3. Critical indices at fluid–fluid phase transition

In this section, we focus attention on the critical indices at fluid–fluid phase transition. Generally, it is accepted that pure fluids at gas–liquid critical points and binary liquid mixtures at liquid–liquid critical points belong to the same universality class as the Ising model [41–45]. For fluid phase transitions driven by short-range interactions, a common scenario is that the Ising criticality exists in the asymptotic region, with a crossover to mean field behavior at large separations from the critical point [46,47]. However, it should be pointed out that the simple power laws involving the universal Ising critical exponents, which are commonly accepted due to the renormalization group theory, are valid only in the so-called asymptotic region near the critical point. Usually, after confirming the validity of the renormalization group theory in such an (narrow) asymptotic critical region, the analysis is extended to a region further from the critical point. Wegner [48] worked out an asymptotic series expansion for critical behavior, and Ley-Coo and Green [49] translated Wegner's expansion into the terminology of fluids. Considering the coexistence curve, as the critical temperature T_c is approached, the difference ΔX of the order parameter (the compositions or densities) in the two coexisting phases vanishes following a scaling law termed the Wegner expansion [47–49]:

$$\Delta X = |X_u - X_l| = B\tau^{\beta}\left(1 + B_1\tau^{\Delta} + B_2\tau^{2\Delta} + \cdots\right). \tag{4}$$

X_u and X_l represent the compositions or densities in the upper and lower phases, respectively. In fluid mixtures, the asymptotic power laws commonly hold in the region $\tau = |T - T_c|/T_c < 10^{-3}$ [47].

The generally accepted universal values of the critical exponents for the 3D Ising-type universality class are $\alpha = 0.11$ for the heat capacity, $\beta = 0.33$ for the coexistence curve, $\gamma = 1.24$ for the compressibility on the critical isochore, and $\delta = 4.8$ for the critical isotherm [50–52]. The corresponding Ising values for various fluid–fluid phase transition are $\alpha = 0.109$, $\beta \sim 0.326$, $\gamma - 1.239$, $\upsilon \sim 0.632$, and $\eta = 0.041$ [52–62]. As mentioned above, the behavior of curves with a power law of $\alpha < 0.2$ cannot be distinguished from the logarithms with $\alpha = 0$ proposed by Zhang [1]. The generally accepted value $\gamma = 1.24$ is very close to Zhang's solution $\gamma = 5/4$. There is a big difference between the generally accepted critical exponent $\beta = 0.326$ for fluid–fluid phase transition (also due to the renormalization group theory) and Zhang's value $\beta = 3/8$. Such difference has been discussed in detail in [1]. Here, it is interesting to know how the generally accepted critical exponent is derived in the literature.

As a routine, one applies first a simple power law, where the exponent β is a free parameter, to fit the experimental data at the asymptotic region near the critical point, and then uses the Wegner-type expansions (i.e., Equation (4)) for the fit with up to two coefficients. In this process, usually, the result ($\beta = 0.326$) of the renormalization group theory is taken as a reference. Then, one discusses the deviations from the asymptotic Ising behavior in terms of Wegner corrections. In this analysis, the exponents are fixed to the universal values $\beta = 0.326$ and $\Delta = 1/2$, while the amplitudes B, B_1, and B_2 are adjustable fitting parameters [47,63,64]. The amplitudes B of the coexistence curve and the amplitudes B_1 and B_2 of the first two Wegner corrections are specific for each system [65]. However, it is difficult to derive conclusions based on the size of the coefficients, because their values change, when the second correction is included into the fit. The statistical error estimated for the parameters becomes unduly large, which shows that the parameters are not independent. It was concluded that the expansion with two correction terms is not appropriate [47]. Furthermore, it would be interesting to see if one could apply a simple power law with Zhang's exponent $\beta = 3/8$ to fit the experimental data over wider asymptotic region near the critical point, and if it would be necessary to fit the critical behavior in terms of Wegner corrections.

Finally, we notice that Monte Carlo studies of the two-component Asakura–Oosawa model give evidence that the critical behavior of colloid–polymer mixtures belongs to the Ising universality class. A linear fit of Verso et al.'s [66] results in the range $-5 \lesssim \log(t) \lesssim -4$ (Figure 12 of [66]) gives a power-law behavior with $\beta = 0.37$, which is very close to Zhang's value $\beta = 3/8$.

4. Conclusion

In conclusion, the critical exponents in some bulk magnetic materials indeed form a 3D Ising universality, which agree well with the recent 3D Ising results derived based on the two conjectures [1]. It is commonly accepted that pure fluids at gas–liquid critical points and binary liquid mixtures at liquid–liquid critical points belong to the same universality class as the Ising model. We suggest to apply a simple power law using Zhang's exponent $\beta = 3/8$ to fit the experimental data over a wider asymptotic region near the critical point of fluid–fluid phase transition.

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