INCOMPLETENESS OF THE CRITICAL EXPONENT DESCRIPTION FOR FERROMAGNETIC SYSTEMS CONTAINING RANDOM IMPURITIES

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We consider the magnetic properties and spin correlation functions of a two-dimensional Ising model with random impurities near the critical temperature and find that the usual parametrization provided by the exponents γ, δ, and η and the correlation length ξ for $T \gtrsim T_c$ does not describe our results. Experimental implications of this breakdown of the critical exponent forms are discussed.

It has become common to analyze experiments on ferromagnetic systems near the Curie temperature in terms of a set of "critical exponents." In particular it is assumed that there exists a unique temperature T_c at which the usually measured macroscopic quantities possess singularities which are commonly parametrized as follows[1]:

$$\text{Specific heat at } H=0 \sim \text{const}(T-T_c)^{-\alpha} \text{ if } T>T_c,$$
$$\sim \text{const}(T_c-T)^{-\alpha'} \text{ if } T<T_c, \text{ or}$$
$$\sim \text{const} \ln|T-T_c|; \tag{1a}$$

$$\text{specific heat at constant } H = C_H \sim \text{const}|H|^{-\zeta}, \text{ or}$$
$$\sim \text{const} \ln|H| \text{ if } T=T_c; \tag{1b}$$

$$\text{spontaneous magnetization} = \lim_{H\to 0+} M(H) = 0 \text{ if } T>T_c,$$
$$\sim \text{const}(T_c-T)^{\beta} \text{ if } T<T_c; \tag{1c}$$

$$\text{zero-field susceptibility} = \left.\frac{\partial M(H)}{\partial H}\right|_{H=0} = \chi(0)$$
$$\sim \text{const}(T-T_c)^{-\gamma} \text{ if } T>T_c,$$
$$\sim \text{const}(T_c-T)^{-\gamma'} \text{ if } T<T_c; \tag{1d}$$

and

$$M(H) \sim \text{sgn}(H)\,\text{const}|H|^{1/\delta} \text{ at } T=T_c. \tag{1e}$$

Furthermore, the correlation between a spin $\sigma(\vec{r})$ at position $\vec{r}$ and a spin $\sigma(\vec{r}\,')$ at position $\vec{r}\,'$ is commonly parametrized when $|\vec{r}-\vec{r}\,'| \to \infty$ and $H=0$ as

$$\langle\sigma(\vec{r})\sigma(\vec{r}\,')\rangle \sim \text{const}|\vec{r}-\vec{r}\,'|^{2-d-\eta} \text{ if } T=T_c, \tag{2a}$$

where d is the dimensionality of the system, and

$$\langle\sigma(\vec{r})\sigma(\vec{r}\,')\rangle \sim M^2(0) + \text{const}|\vec{r}-\vec{r}\,'|^{-a} e^{-|\vec{r}-\vec{r}\,'|/\xi} \text{ if } T \neq T_c, \tag{2b}$$

where

$$\xi = \text{const}(T-T_c)^{-\nu} \text{ if } T>T_c,$$
$$= \text{const}(T_c-T)^{-\nu'} \text{ if } T<T_c, \tag{2c}$$

and a may be different if T is above or below T_c. In (2a) and (2b) the "constants" may depend on the angle between $\vec{r}$ and $\vec{r}\,'$.

The only compelling reason to believe that these specific parametrizations should be sufficient to describe magnetic critical phenomena is that for simplified models in which some of the quantities in (1) and (2) can be analytically studied, the "critical-exponent" form has been found to hold. The purpose of this note is to show that these forms cannot be universally valid, by discussing a model whose critical behavior is not described by (1a), (1d), (1e), and (2). Such breakdowns of "critical-exponent" forms are particularly significant since apparent violations of (1a) and (1b) have been observed in several recent experiments.[2]

The model most commonly known to obey the "critical-exponent" forms is Onsager's two-dimensional Ising lattice whose horizontal bonds are all of strength E_1 and whose vertical bonds are all of strength E_2. The model we consider is a modification of Onsager's lattice which keeps all horizontal bonds E_1 the same and keeps all vertical bonds $E_2(j)$ between rows j and $j+1$ the same but lets $E_2(j)$, $j=\cdots, 1, 2, \cdots$, be independent random variables with a distribution of narrow width w. The specific heat at $H=0$ of this model has been previously calculated.[3] It was found that when $1-T/T_c=O(w^2)$, $C_{H=0}$ deviates drastically from the logarithimic divergence of Onsager's lattice and at T_c has a singularity that is infinitely differentiable [and hence is not of the form (1a)]. The investigation of the remaining quantities in (1) and (2) has been done in two steps:

(I) We have considered this random two-dimensional Ising model on a half plane (the rows are numbered $j=1, 2, \cdots$) and have investigated the boundary magnetization $M_1(\mathcal{H})=\langle\sigma(k,1)\rangle$ and the spin-spin correlation function for two spins in the boundary row $S_{1,1}(N,\mathcal{H})=\langle\sigma(0,1)\sigma(N,1)\rangle$. The boundary row is in the horizontal direction so the bonds in this row are all E_1. The magnetic field $\mathcal{H}$ is allowed to interact solely with the boundary row. The functions M_1 and $S_{1,1}$ are not the same for all sets $\{E_2\}$. Therefore we have computed their average values when averaged over all sets of energies $\{E_2\}$. We find that when $|1-T/T_c|=O(w^2)$ and $|\mathcal{H}|=O(w)$ there are significant deviations from the "critical-exponent" forms that describe this Ising model when $|1-T/T_c|\gg w^2$ or $|\mathcal{H}|\gg w$. The details of the calculations are lengthy and are reported elsewhere.[4] The major results are as follows:

(1) There exists a range of temperatures $-T_{(1)}<T_c<T_{(1)}$ in which the average zero-field boundary susceptibility is infinite. There exists a still larger range about T_c in which $\langle\chi(0)\rangle_{\rm Av}$ exists but $\langle M_1(\mathcal{H})\rangle_{\rm Av}$ is not analytic at $\mathcal{H}=0$.[5]

(2) When $T=T_c$ and $|\mathcal{H}|\ll w$,

$$\langle M_1(\mathcal{H})\rangle_{\rm Av}\sim -C_1\,{\rm sgn}(\mathcal{H})w\ln[|\mathcal{H}|/wkT_c]^{-1}, \tag{3}$$

where k is Boltzmann's constant and C_1 is known.

(3) When $T=T_c$, $\mathcal{H}=0$, and $N\gg w^{-2}$,

$$\langle S_{1,1}(N,\mathcal{H})\rangle_{\rm Av}\sim C_2w^{-2}[\ln w^2N]^{-1}, \tag{4}$$

where C_2 is known.

(4) When $\mathcal{H}=0$, $T-T_c=O(w^{-2})$, and $N\gg w^{-2}$, $\langle S_{1,1}(N,\mathcal{H})\rangle_{\rm Av}$ approaches $\langle M_1^2(0)\rangle$ as an inverse power of N. The precise value of the power depends on $T-T_c$.

(5) When $T\to T_{c-}$,

$$\langle M_1(0)\rangle_{\rm Av}\sim C_3(T-T_c), \tag{5}$$

where C_3 is known. This is to be compared with

$$M_1(0)\sim \bar{C}_3(T-T_c)^{1/2} \tag{6}$$

which holds for the corresponding Onsager lattice with the same E_1 and T_c. In the above, T_c is that temperature at which $C_{H=0}$ was found[3] to be nonanalytic.

These results may be summarized by saying that $\langle M_1\rangle_{\rm Av}$ and $\langle S_{1,1}(N,\mathcal{H})\rangle_{\rm Av}$ are not describable by critical exponents γ, γ', δ, and η, and the correlation length ξ.

(II) The second step is to use Griffiths' theorem[6] to show that $\langle M_1(\mathcal{H})\rangle_{\rm Av}$ and $\langle S_{1,1}(N,\mathcal{H})\rangle_{\rm Av}$ are lower bounds on the corresponding bulk properties. Such a bound exists because for any Ising model on the full plane ($j=\cdots, 0, 1, 2, \cdots$) whose interaction energies are all non-negative, we cannot increase the value of $\langle\sigma(k,1)\rangle$ and $\langle\sigma(0,1)\times\sigma(N,1)\rangle$ by (1) reducing H to zero except where H interacts with a spin in row 1 and (2) decreasing the bonds $E_2(0)$ between row 0 and row 1 to zero. Since this remark holds for every set of $E_2(j)$, we may average the resulting inequality over all sets of $E_2(j)$. Furthermore, $M(H)$ in the bulk is, with probability one, the same for all sets $\{E_2\}$, so we find

$$\langle M_1(\mathcal{H})\rangle_{\rm Av}\leq\langle\langle\sigma(k,1)\rangle\rangle_{\rm Av}=M(H) \tag{7}$$

and

$$\langle S_{1,1}(N,\mathcal{H})\rangle\leq\langle\langle\sigma(0,1)\sigma(N,1)\rangle\rangle_{\rm Av} =\langle\langle\sigma(0,k)\sigma(N,k)\rangle\rangle_{{\rm Av\ over\ }k}. \tag{8}$$

These lower bounds may be used to draw conclusions about the critical behavior of the bulk properties if we make the assumption that the temperature T_c at which $C_{H=0}$ was shown to fail to be analytic is the same as the temperature at which $M(0)$ vanishes. While there exists no proof that these two temperatures must be the same, neither does a counter example exist. Indeed, if this connection between the disappearance of the order parameter $M(0)$ and the singularity in $C_{H=0}$ is removed it becomes difficult to see why $C_{H=0}$ should not be analytic at all temperatures and the critical-exponent forms (1) and (2) clearly require modification. Therefore, we make this assumption and find that result (5) is a bound on the rate at which $M(0)$ vanishes as $T \to T_{c-}$ and that results (1)-(4) are lower bounds on the corresponding bulk properties if $T \geq T_c$. These results may be summarized by saying that the "critical exponents" γ, δ, and η, and the correlation length ξ for $T \gtrsim T_c$ do not exist for our model.

It is perhaps not surprising that "critical exponents" do not provide an adequate description of this model of immobile random impurities. This "critical-exponent" description has been abstracted from studies of pure materials of infinite size where the correlation length ξ is the only relevant length scale. In a system containing immobile random impurities there exists a second length scale that measures the effectiveness of the impurities in modifying the correlations in the pure material. This length must go to infinity as $w \to 0$. However, for w fixed, if T is sufficiently close to T_c and H is sufficiently close to zero such that ξ, the "critical-exponent" correlation length, is comparable with the impurity-length scale but still small compared with the size of the sample, the effect of even a small amount of impurities may no longer be ignored.

At present there is no compelling reason to assume that when ξ is much smaller than the impurity-length scale the "critical-exponent" forms fail to describe the behavior of real ferromagnets. However, when ξ becomes comparable with the impurity length our model suggests that there are three ways in which the observed forms (1) may change:

(1) Decrease to smooth behavior. A specific heat that has an infinitely differentiable essential singularity at T_c can never be distinguished experimentally from form (1a) with α and α' large and negative. Any quantity that behaves smoothly cannot be used for an unambiguous measurement of T_c.

(2) Maintain an observable "critical exponent." The experimental distinction between classes 1 and 2 is obviously somewhat imprecise. In this model if the spontaneous magnetization is describable by (1c) then β must be positive and less than 1. However, because of the possibility of having infinite zero-field susceptibility near T_c, caution must be exercised in making sure that H is close enough to zero so that a measurement of the "spontaneous" magnetization will yield the true value.

(3) Increase to more singular behavior. From this model calculation we speculate that in real magnetic systems at T_c when $H \sim 0$, $M(H)$ will be larger than any power law permits. Furthermore, the zero-field susceptibility may diverge at a temperature other than T_c so that it may be impossible to use the susceptibility to locate the Curie temperature.

These considerations indicate that the critical behavior of systems containing immobile random impurities is considerably more complex than that indicated by the "critical-exponent" forms. Furthermore, these model calculations serve to make more concrete the somewhat vague notion that impurities will "smear out" the phase transition. At present, no measurements of any property other than the specific heat has been made on a ferromagnetic system whose specific heat deviates from (1a) and (1b) for T sufficiently close to T_c and H sufficiently close to zero in the range of T and H where these specific-heat deviations occur. Experimental studies of these other properties listed in (1) and (2) should prove to be an extremely fruitful area for future investigations into critical phenomena.

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[1]See for example, L. Kadanoff, W. Götze, D. Hamblen, R. Hecht, E. A. S. Lewis, V. V. Palciauskas, M. Rayl, F. Swift, D. Aspnes, and J. Kane, Rev. Mod. Phys. 39, 395 (1967). The form (1b) is not mentioned here. For this form we use Ref. 2.

[2]B. J. C. van der Hoeven, D. T. Teaney, and V. L. Moruzzi, Phys. Rev. Letters 20, 719 (1968). This is the only measurement of a specific heat near T_c that studies the dependence on both T and H. There are several other experiments on both ferromagnets and antiferromagnets, however, which have revealed deviations from the form (1a) for $T/T_c - 1$ sufficiently small. For example, Ni was studied by P. Handler, D. E. Mapother, and M. Rayl, Phys. Rev. Letters 19, 356 (1967); $RbMnF_3$ was studied by D. T. Teaney, V. L. Moruzzi, and B. E. Argyle, J. Appl. Phys. 37, 1122 (1966); and dysprosium aluminum garnet was studied by B. E. Keen, D. P. Landau, and W. P. Wolf, J. Appl. Phys. 38, 967 (1967). It must be emphasized that in these experiments critical exponents do describe the specific heat for T not too close to T_c or H not too close to zero. This should be distinguished from the well-known case of alloys of a small concentration of Fe in a large concentration of Pd where the measurements of B. W. Veal and J. A. Rayne, Phys. Rev. 135,

A442 (1964), indicate that at $H=0$ there is no temperature range in which the specific heat has a singularity of the form (1a) with a measurable α.

[3]B. M. McCoy and T. T. Wu, Phys. Rev. 176, 631 (1968), and Phys. Rev. Letters 21, 549 (1968).

[4]B. M. McCoy, to be published. This work relies on that of B. M. McCoy and T. T. Wu, to be published.

[5]A related lack of analyticity of the bulk magnetization at $H=0$ has been discovered in another random two-dimensional Ising model by R. Griffiths, Phys. Rev. Letters 23, 17 (1969).

[6]R. Griffiths, J. Math. Phys. 8, 478 (1967).

THRESHOLD PHOTONEUTRON CROSS SECTION FOR Mg^{26} AND A SOURCE OF STELLAR NEUTRONS*

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The differential photoneutron cross section for Mg^{26} at 135° has been measured as a function of photon energy from 10 keV to 1.5 MeV above threshold by the threshold photoneutron technique. Several prominent resonances have observed, including one located at 54.3 keV above threshold. The existence of this resonance in the Mg^{26} compound system might provide, through the reaction $Ne^{22}(\alpha, n)Mg^{25}$, the primary production mechanism for neutrons in stars.

Extensive studies on stellar processes have produced strong evidence that neutron capture plays the dominant role in the synthesis of elements heavier than iron.[1,2] However, the specific nature of the source of the neutrons needed for these processes still is uncertain. The most likely sources appear to be the reaction[1,3,4] $Ne^{21}(\alpha, n)Mg^{24}$ and the reaction[5] $Ne^{22}(\alpha, n)Mg^{25}$ (see also Reeves[6] and Peters[7]). This paper reports the discovery of a resonance in the Mg^{26} compound system which might enhance significantly the neutron production from the latter reaction.

The resonance was observed with the threshold photoneutron technique. This technique, which has been described in detail elsewhere,[8,9] was used to measure the photoneutron cross section for Mg^{26} just above the neutron separation energy of 11.1 MeV. It consists of a high-resolution time-of-flight measurement of the spectrum of neutrons photoejected from a nuclear sample by a bremsstrahlung beam, when the bremsstrahlung end-point energy is limited so that the state of the residual nucleus is known. The sample consisted of 85.5 g of MgO enriched to 99.7% Mg^{26}. The $Mg^{26}(\gamma, n)Mg^{25}$ cross-section data presented here were taken at an electron beam energy of 13.3 MeV and with an angle of 135° between the incident beam direction and the neutron flight tube. Additional measurements were made at several lower beam energies to measure background and to determine the correspondence between the peaks in the photoneutron spectrum and the states of the residual nucleus. Also, a measurement was made at 90° in order to help identify the spin and parity assignments of the prominent levels. A new neutron detector[10] was used, located 17 m from the sample. Its efficiency ranges from about 12% at 2 keV to 4% at 2 MeV, and its response time was comparable with the 30-nsec beam-burst width.

The measured differential photoneutron cross section at 135° as a function of photon energy E_γ and of laboratory neutron energy E_L is shown in Fig. 1. The energies of the peaks of the most prominent resonances are at $E_L = 54.3$, 63.2, 182, 224, 358, 392, 617, and 1109 keV. Peak energies that correspond to neutron emission to the first excited state (rather than the ground state) of Mg^{25} occur at $E_L = 432$ and 737 keV. The areas under the curves for the resonances whose peaks are at $E_L = 617$ and 1109 keV ($E_\gamma = 11.752$ and 12.268 MeV, respectively) are identical to those obtained with monoenergetic photons by Fultz et al.[11] when the angular distribution appropriate to $E1$ photon absorption (followed by p-wave neutron emission) is used. For the very prominent resonance at $E_L = 54.3$ keV, the ground-state γ-ray width $\Gamma_{\gamma 0}$ measured in the present experiment and extracted by area analysis is 1.75 eV, with an uncertainty no greater than 10%. (The monoenergetic photon measurement[11] is accurate to within 7%.)

The astrophysical importance of this work lies in the discovery of the resonance at 54.3 keV, provided that its spin and parity allow the α-particle capture to proceed ($J^\Pi = 0^+, 1^-, 2^+, \cdots$, the "natural-parity" states). Since zero-spin levels in Mg^{26} cannot be excited by real photons from