Equilibrium Properties of Ferromagnets and Antiferromagnets in the Vicinity of the Critical Point*

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Within the past few years there has been a considerable advance both theoretically and experimentally in our knowledge of the behavior of magnetic systems in the vicinity of the critical temperature. The present paper consists of a presentation of some of the new experimental information and its connection with the new theoretical predictions.

Some of the equilibrium properties of magnetic systems which are important in the description of a ferromagnet in the critical region are the following:

1. The spontaneous magnetization versus temperature in zero external field,
2. the magnetic susceptibility $(\partial M/\partial H)_{H=0}=\chi$,
3. the magnetization versus field at the critical temperature T_c,
4. the specific heat at constant volume C_v, and
5. the thermal expansion coefficient.

In the discussion which follows, we shall present the experimental data and theoretical predictions now available on each of the properties listed above. In view of the fact that each of these properties has an analog in the liquid-gas system, as has been emphasized by Fisher [1] and his coworkers [2], we shall make the connection between each magnetic property and its fluid analog.

1. *The spontaneous magnetization.* In figure 1 we present a schematic drawing of the temperature dependence of the spontaneous magnetization $M(T)$ of a ferromagnet normalized to its value at $T=0$, $(M(0))$, in zero external field $(H=0)$. Along side this we present the fluid analog, namely the temperature dependence of the liquid-gas density along the coexistence curve. In the critical region the magnetization and orthobaric density data can be fit to equations of the form:

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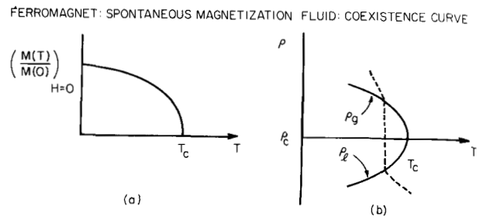

FIGURE 1. (a) *A schematic plot of the temperature dependence of the magnetization of a ferromagnet.* (b) *The fluid analog: the liquid and gas densities along the coexistence curve.*

$$\left(\frac{M(T)}{M(0)}\right)_{H=0} \rightrightarrows D\left(1-\frac{T}{T_c}\right)^{\beta} \tag{1}$$

$$\left(\frac{\rho_l-\rho_g}{2\rho_c}\right) \rightrightarrows D'\left(1-\frac{T}{T_c}\right)^{\beta'}. \tag{2}$$

The first experimental measurements of D and β were reported in 1962 by P. Heller and G. Benedek [3] through a study of the nuclear resonance of the F^{19} nucleus in the antiferromagnet MnF_2. In table 1 we show the temperature range $\Delta T/T_c$ over which (1) was found to fit the data, and the corresponding values of D and β. Equation (1) holds in MnF_2 over a temperature range $7\times10^{-5} \leqslant (\Delta T/T_c) < 0.1$. The closest point measured was about 3 mdeg from the critical point. This data shows that $\beta=0.333\pm0.003$. This result is in disagreement with molecular field theories which give $\beta=\frac{1}{2}$ in the limit $T\rightarrow T_c$. In view of the marked disparity between this result and that of the classical theories of magnetism, Heller and Benedek [4] later carried out a careful investigation of the insulating ferromagnet EuS. This crystal

TABLE 1. *Experimental and theoretical values for the parameters describing the temperature dependence of the spontaneous magnetization of a ferromagnet or antiferromagnet*

	$\left(\frac{\Delta T}{T_c}\right)$	D	β
MnF_2	$0.10 \leftrightarrow 7\times10^{-5}$	1.200 ± 0.004	0.333 ± 0.003
EuS	$0.08 \leftrightarrow 1\times10^{-2}$	1.145 ± 0.02	$0.33\ \pm0.015$
$CrBr_3$	$0.04 \rightarrow 0.7\times10^{-2}$	1.32 ± 0.07	0.365 ± 0.015
Molecular field theory		1.48 (S=5/2) 1.44 (S=7/2)	0.500
Ising model		1.49	0.3125

is cubic, the magnetic europium spins are localized on the lattice sites and each magnetic ion has zero orbital angular momentum and S=7/2. This crystal represents the closest known physical approximation to the ideal "Heisenberg" ferromagnet. The results of this investigation are summarized in table 1. The one-third power law was found to apply also to this system.

It is worth noting that the 1/3 power law starts to fit the data on EuS and MnF_2 when $\Delta T/T_c \sim 9$ percent. The magnetization at this point is about 1/2 the saturation value. Thus the 1/3 power law applies to the magnetization data over a rather wide range in M, namely $1/2 > (M(T)/M(0)) \geq 0$. In row 3 of table 1 we also present new measurements of D, β and $\Delta T/T_c$ made very recently by S. D. Senturia at MIT on the ferromagnetic insulator $CrBr_3$. This material is particularly interesting from the theoretical point of view because the ferromagnetic chromium spins are arranged in planes, and each plane is separated from the next plane by a plane of nonmagnetic Br ions. As a result, the coupling between the magnetic spins is very anisotropic. The exchange constant for coupling in the plane is 16 times stronger than the constant for coupling between the planes. Thus, this ferromagnet is almost two-dimensional, with only a small three-dimensional admixture. Nevertheless, this system behaves somewhat like the ideal three-dimensional Heisenberg ferromagnet EuS in that $\beta=0.365$. The critical region for $CrBr_3$ sets in a $\Delta T/T_c \sim 5\times10^{-2}$ at which point $M(T)/M(0) \sim 0.41$.

In table 1 we also present the theoretical predictions of the Ising [5] and molecular field models. No theoretical prediction has yet appeared for the Heisenberg model. It should also be mentioned that H. Callen and E. Callen [6] have calculated $M(T)$ theoretically using a sophisticated cluster approximation, and report that $M(T)$ follows the 1/3 power law provided that T is not too near T_c. In the limit $T\rightarrow T_c$, of course, their cluster theory goes over to the $\beta=1/2$ value.

As an example of the analogous behavior of a fluid we point out the work of Weinberger and Schneider [7, 1]. They found that eq (2) fits the data on xenon for $\Delta T/T_c$ as small as 3×10^{-5}, and that $\beta'=0.345\pm0.015$ *and* $D'\simeq1.61$. The van der Waals equation gives $\beta=1/2$. In fact, Landau's theory [8] of the critical region shows that $\beta=1/2$ follows from the very general assumption that the free energy can be expanded as a power series in density and temperature around the critical point.

Thus the breakdown of this law reflects a rather unusual mathematical and physical character of the critical point.

From an historical standpoint it should be noted that the departure of β from the classical value for magnetic systems was discovered at about the same time that important theoretical advances were being made. In 1961 and 1962 Domb and Sykes [9, 10] were able to show from a study of their high temperature expansions that the susceptability of a ferromagnet in the Ising and Heisenberg models did not diverge as slowly as was expected by the molecular field theories. Also, Baker [11, 12] introduced at this time the method of Pade approximants as an accurate and reliable means of extrapolating the high temperature expansions into the critical region. This extrapolation showed [13] that the susceptibility as calculated on the Heisenberg and Ising Hamiltonians diverged more rapidly than that predicted by the molecular field theories. These developments and the observation, which was stressed particularly by M. E. Fisher, of the close similarity in the behavior of systems as disparate as the ferromagnet and the fluid greatly contributed to the present expansion of work in this field.

2. *The susceptibility.* In figure 2 we show the temperature dependence of the isothermal susceptibility of a ferromagnet as $T \to T_c$. Along side it is shown the temperature dependence of the analogous property of the fluid, the isothermal bulk modulus along the critical isochore. The susceptibility of the ferromagnet is defined as

$$\chi = (\partial M(T, H)/\partial H)_{H=0}. \tag{3}$$

As one approaches T_c from the high temperature or paramagnetic side, χ diverges like

$$\chi \underset{(T \to T_c)}{=} C/(1 - T/T_c)^{\gamma}. \tag{4}$$

As one approaches T_c from the ferromagnetic side, χ can be fit to an equation of the same form as (4) but one must leave the possibility that the coefficient D and exponent γ have different values for $T < T_c$. Thus, on this side of T_c we write

$$\chi \underset{(T \to T_c)}{=} C'/(1 - (T/T_c))^{\gamma'}. \tag{5}$$

In the fluid the analog is the isothermal compressibility β_T.

$$\beta_T = \frac{1}{\rho}\left(\frac{\partial \rho}{\partial P}\right)_T. \tag{6}$$

This can also be written in terms of the chemical potential μ using the thermodynamic identity that along an isotherm

$$\left(\frac{\partial P}{\partial \mu}\right)_T = \rho. \tag{7}$$

Thus

$$\beta_T = \frac{1}{\rho^2}\left(\frac{\partial \rho}{\partial \mu}\right)_T. \tag{8}$$

As $T \to T_c$ from above, the compressibility at $\rho = \rho_c$ diverges as follows:

$$\beta_{T, \rho_c} \underset{(T \to T_c)}{=} d/(1 - (T/T_c))^{\Gamma}. \tag{9}$$

Below T_c the relevant compressibility is that evaluated right at the coexistence line for either the liquid or the gas

$$\beta_{T, \rho_c \text{ or } \rho_{\ell g}} \underset{(T \to T_c)}{=} d'/(1 - (T/T_c))^{\Gamma'}. \tag{10}$$

We see that in the liquid case, the pressure, or the chemical potential plays the same role as does the magnetic field in the ferromagnetic case.

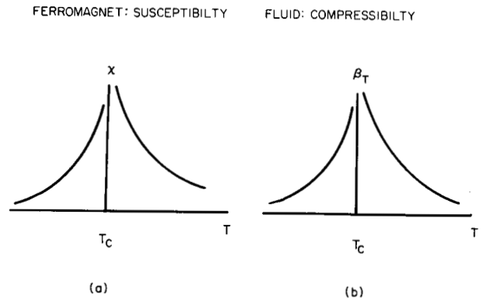

FIGURE 2. (a) *The temperature dependence of the isothermal susceptibility of a ferromagnet as* $T \to T_c$. (b) *The temperature dependence of the isothermal compressibility of a fluid evaluated at* $\rho = \rho_c$ *for* $T > T_c$ *and at* $\rho = \rho_l$ *or* $\rho = \rho_c$ *for* $T < T_c$.

The first comparison between the new theoretical predictions [9, 10, 13, 14] and experiments was provided by the work of Jacrot [15] who determined γ through a study of the scattering of neutrons from iron as $T \to T_c$ from above. His measurements indicated that $\gamma = 1.3$. Stimulated by the departure of γ from the classical value $\gamma = 1$, Noakes and Arrott [16] reexamined their older measurements of the susceptibility of iron for $T > T_c$, and they also carried out new measurements of this quantity. They found $\gamma = 1.37 \pm 0.04$. This result is tabulated in table 2 where we give γ for each ferromagnet which has been studied up till now. We also give in column 2 the maximum range over which eq (4) actually fits the experimental data. The range is $0 < \Delta T/T_c < \Delta T_M/T_c$, and $\Delta T_M/T_c$ represents the outer limit of the critical region insofar as the susceptibility is concerned. In their paper Noakes and Arrott pointed out that earlier data by S. Arajs [17], when analyzed in terms of eq (4) also gave a nonclassical value for γ. This observation was later confirmed by Arajs [18] who has measured γ from his studies on Fe, Co, and Ni. His important results on γ and $\Delta T_m/T_c$ are given in table 2. Kouvel and Fisher also examined the divergence in the susceptibility of nickel by going back to the very early careful work of P. Weiss and R. Forrer [20]. This gave $\gamma = 1.35 \pm 0.02$. Graham [21] has studied the susceptibility of Gd above the Curie point and concludes $\gamma = 1.3$ but does not give error limits for this estimate. Furthermore, his data indicates that γ depends on whether the magnetic field is applied along the c-axis or perpendicular to it.

In table 2 we also list in the last three rows the values of γ as obtained theoretically using the molecular field, Ising and Heisenberg models along with literature references to these calculation.

Table 2. *Experimental and theoretical values for the parameters describing the temperature dependence of the susceptibility of a ferromagnet as* $T \to T_c$ *from above*

Ferromagnet	$(\Delta T_M/T_c)$	γ	References
Fe	3×10^{-2}	1.3	15, 13
		1.37 ± 0.04	16
		1.33 ± 0.04	18
Co	1×10^{-2}	1.21 ± 0.04	18
Ni	2×10^{-2}	1.35 ± 0.02	19
		1.29 ± 0.03	18
Gd	4×10^{-2}	1.3	21
Theory			
Molecular field		1.00	
Ising		1.25	9, 11
Heisenberg		1.33 ± 0.01	13, 14

It is worth stressing that we do not know of data on γ' the parameter which describes the divergence of χ as $T \to T_c$ from the ferromagnetic side of the Curie point.

In the case of a fluid, the classical theories give $\Gamma = 1.0$. Because of the experimental difficulties, data on the isothermal compressibility appears to be scanty and insufficiently precise in the critical region. The measurements of Habgood and Schneider [22] indicate only that [1] $\Gamma > 1.1$. The Ising theory for the liquid predicts $\Gamma = 1.25$.

3. *The critical isotherm.* The next equilibrium property of a ferromagnet which is of particular interest in the critical region is the dependence of M on H at $T = T_c$. The divergence of the susceptibility as $T \to T_c$ shows that M is an extremely strong function of H at T_c. In figure 3 we show a schematic plot of M versus H at T_c. Along side it we present the fluid analog, namely the dependence of $(\rho - \rho_c)$ on the pressure for $T = T_c$, i.e., the shape of the critical $\rho - p$ isotherm. The data on both these systems may be fit to equations of the following form:

$$M(T_c, H) = AH^{1/\delta} \tag{11}$$

and

$$(\rho - \rho_c) \underset{T=T_c}{=} A'(p - p_c)^{1/\delta'} = A''(\mu - \mu_c)^{1/\delta'} \tag{12}$$

where μ is the chemical potential. The index δ or δ' is a measure of the verticality of the critical isotherms as sketched schematically in figure 3, as these indices are measures of the lowest nonvanish-

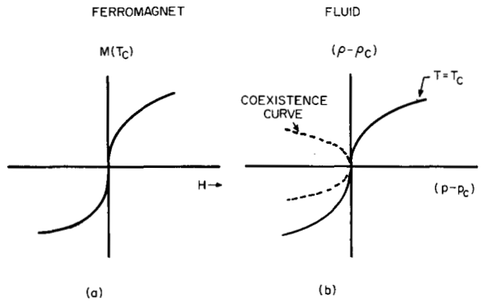

FIGURE 3. (a) *The dependence of the spontaneous magnetization* M *on the magnetic field* H *at the Curie point* ($T = T_c$). (b) *The shape of the critical isotherm of a fluid.*

ing derivative of H (or μ) relative to M (or ρ). In magnetic systems δ has been found for two materials, nickel and gadolinium. The information on nickel was obtained from Weiss and Forrer's 1926 measurements [20] by Kouvel and Fisher [19]. They found $\delta = 4.22 \pm 0.04$. Graham [21] reports that his studies of gadolinium give $\delta = 4$, without placing error limits on this figure. The molecular field theory predicts $\delta = 3$ while the best present analysis of the Ising model in three dimensions strongly indicates that the value $\delta = 5.2$ is the correct theoretical value for this Hamiltonian. There are as yet no calculations of δ using the Heisenberg Hamiltonian. These results are collected in table 3.

In the fluid there is considerably more data [23] on the shape of the critical isotherm. The data on Xe, Co_2, and H_2 indicates [2, 24] that for the fluid $\delta' = 4.2 \pm 0.1$. Once again we see the close similarity in behavior between the ferromagnet and the fluid. Theoretically the van der Waals theory, which is the analog of the molecular field theory of ferromagnetism, predicts a cubic equation for the $\rho - p$ critical isotherm, i.e., $\delta' = 3$. The Ising model as analyzed by Gaunt, Fisher, Sykes, and Essam [2] gives $\delta' = 5.2$. We tabulate these results in table 4.

4. *The specific heat.* Of all the physical quantities that vary rapidly near the critical point, the specific heat appears to diverge most delicately. Onsager's theoretical treatment [25] of the two-dimensional Ising system indicates that the specific heat diverges logarithmically as $T \to T_c$. Such a slow divergence requires very high precision in the measurement of the specific heat, and accurate temperature control very near to the critical point. A very careful study of the specific heat of a magnetic system has recently been carried out by Skalyo and Friedberg [26] on the antiferromagnet $CoCl \cdot 6H_2O$. These workers find that as one approaches T_c from above (+) or from below (−) that the specific heat appears to diverge like

$$\left(\frac{C_p(T)}{R}\right)^{\pm}_{H=0} = -a \ln \left| 1 - \frac{T}{T_c} \right| + \Delta_{\pm}. \tag{13}$$

This behavior begins when $(T - T_c)/T_c$ is of the order of a few percent. However, in the immediate vicinity of T_c ($(T - T_c)/T_c \sim -5 \times 10^{-3}$ and $+1 \times 10^{-3}$) this divergence does not continue. Presumably this is due to the presence of crystal strains and the resulting coexistence of the antiferromagnetic and paramagnetic phases.

TABLE 3. *Experimental and theoretical values of parameters describing the shape of the critical isotherm for a ferromagnet*

Ferromagnet	δ	Reference
Ni	4.22 ± 0.04	19
Gd	4	21
Theory		
Molecular field	3.0	
Ising	5.2 ± 0.15	2

TABLE 4. *Experimental and theoretical values of parameters describing the shape of the critical isotherm for a fluid*

Fluid	δ	References
CO_2, H_2, Xe	4.2 ± 0.1	2, 23, 24
Theory		
van der Waals	3.0	
Ising	5.2 ± 0.15	2

In addition, Yamomoto, et al. [27] have recently surveyed the literature for specific heat measurements on magnetic systems. They fit the experimental data to a logarithmic divergence of the form of eq (13), and present a table listing the resulting values of a and $(\Delta_+ - \Delta_-)$ for 28 ferromagnetic and antiferromagnetic elements and compounds.

It may be well to stress that while the data does show that C_p does get large very slowly as $T \to T_c$ the range over which the logarithmic dependence applies is $\sim 5 \times 10^{-3} < (T - T_c)/T_c \lesssim 5 \times 10^{-2}$. Measurements examining the question of continuance of the logarithmic divergence closer to the critical point are needed. Along these same lines it should be pointed out that what is measured experimentally is C_p and what is calculated theoretically is C_v. The difference is proportional to α^2/β_T where α is the thermal expansion and β_T

the isothermal compressibility. It is known that the thermal expansion increases markedly as $T \to T_c$, but accurate data is not now available giving the precise behavior of $\alpha(T)$ and $\beta(T)$. In view of the slow divergence of C_p it is particularly necessary to determine the importance of the $C_p - C_v$ correction term before reaching conclusions about the precise mathematical nature of the divergence of C_v.

In the case of the fluid, while the van der Waals theory predicts no divergence in C_v as $T \to T_c$, the experimental data has indicated [23] that C_v along the critical isochore grows very large as $T \to T_c$. Recently the work of Voronel and his coworkers [28, 29] has shown that the specific heat at constant volume in argon and oxygen apparently diverges logarithmically as $T \to T_c$. The data of these workers has recently been very carefully examined and discussed in the light of the best present theoretical information [30, 31].

Conclusions

Since 1961–1962 a considerable amount of experimental data on the equilibrium properties of magnetic systems has been obtained. The behavior of each of these properties in the critical region is in striking and clear-cut disagreement with the predictions of the "classical" molecular field theories of ferromagnetism. The development of the method of Pade approximants, in combination with the high temperature expansions of the partition functions have enabled calculations of the equilibrium properties of the ferromagnet using the Ising Hamiltonian. The results of such calculations are closer to the data than the "classical" theories, however, by and large the experimental results indicate that the Ising model is inadequate to describe a real ferromagnet. The Heisenberg Hamiltonian has been used successfully to calculate the divergence of the susceptibility and the results are in very good agreement with the experiment.

At this point in the development of the subject certain directions for the future work seem clear. On the experimental side there is a need for much more information particularly on the shape of the critical isotherm, the specific heat, the thermal expansion, and the susceptibility below T_c. Also, we have to have more data on the localized spin systems rather than the ferromagnetic metals. In the insulators we may expect that the Heisenberg Hamiltonian will apply, while the appropriate Hamiltonian for the metals is not known. Also, it would be most desirable to measure several equilibrium properties on a single ferromagnet.

On the theoretical side, the clear need is a calculation of the magnetization, the critical isotherm, the susceptibility below T_c and the specific heat using the Heisenberg Hamiltonian. And finally, the close analogy between the fluid and the ferromagnet, combined with the clear breakdown of the conventional expansions of the free energy in a power series around T_c suggest that the behavior of three-dimensional ferromagnets and fluids near the critical point may be a result of some very general property connected with the ordering and is relatively independent of the details of the Hamiltonian of the system.

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Experimental Studies of Magnetic Ising Systems Near the Critical Point*

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The Ising model has long been used as a convenient approximation for the investigation of magnetic order-disorder transistions, but until recently no real magnetic substances were known in which the assumptions of the model coincided with reality. The advent of many new rare earth compounds has changed this position. There are now compounds in which it is an excellent approximation to treat the magnetic ions as effective spins $s' = 1/2$, whose magnetic moments $\mu = g \cdot s'$ are constrained to one direction by an extremely anisotropic g tensor [1]. A simple material of this kind is dysprosium aluminum garnet (DAG) which orders antiferromagnetically at about 2.5 °K. The properties of DAG in the critical region have been investigated by a number of workers over the past three years [2–9].

The principal difference between such a material and the Ising model lies in the importance of long range magnetic dipole forces, which account for a major fraction of the total interactions. Dipolar forces lead to a shape dependence of all the magnetic properties (even the "zero field" susceptibility) and at present there is no theory better than the molecular field to allow for these effects. With a molecular field calculated from suitable lattice sums, it is possible to remove most of the long range effects, and to reduce measured magnetic properties to those corresponding approximately to a material with nearest neighbor forces only [7]. This has been done for the susceptibility of DAG in the region of the critical point. It was found that the results could be fitted to law of the form predicted by the Ising model theory [10]:

$$\chi T = \lambda [\xi_c - B_{\pm}, (1 - T/T_c) \log |1 - T/T_c|],$$

with λ equal to the known Curie constant, 10.35/mol and ξ_c, B_+, and B_- respectively 0.565, 0.30, and 0.80. $T_c = 2.493$ °K is critical (Néel) temperature in zero field. The values for ξ_c, B_+, and B_- are not strictly comparable with those given by any of the theoretical estimates since none of these have considered the garnet structure explicitly, but it is interesting to note that the ratio $B_-:B_+$ is close to the value 3 found for other three dimensional structures.

The "reduced" susceptibility measurements have also been used to compare Fisher's predicted relation for the specific heat [11], $C = A \frac{\partial}{\partial T} (\chi T)$, with the observed values [7]. (A is a slowly varying function of temperature whose value at T_c can be estimated from other measurements.) Excellent agreement was found over a range of temperatures $0.5\ T_c < T < 1.5\ T_c$.

Critical effects at temperatures below T_c have also been investigated as a function of magnetic field [4]. The most striking result is that the transition from the antiferromagnetic state to the paramagnetic state seems to be first order for $T < \sim 0.6\ T_c$ and higher order for $\sim 0.6\ T_c < T < T_c$ [12]. A first order transition is not predicted by any of the simple Ising models at any finite temperature, and it is probably a result of the long range interactions.

Accompanying the change from antiferromagnetic to paramagnetic order in an applied field there are magneto-caloric effects and these have been studied by measuring adiabatic changes of temperature with field [13]. In contrast to the isothermal transitions, which are completely reversible, the adiabatic changes showed a large and as yet unexplained hysteresis.

Although most of the critical point measurements on Ising-like systems have so far been concentrated on dysprosium aluminum garnet, there is reason to believe that other rare earth compounds will be found which also approximate to an Ising model [14]. It seems rather certain that almost all materials of this kind will only order at low temperatures, where long-range magnetic dipole interactions are important, and it would therefore be very helpful

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