MAGNETIC EQUATION OF STATE FOR NICKEL NEAR ITS CURIE POINT

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The magnetization-field-temperature (σ-H-T) behavior of nickel both above and below its Curie point T_C is found to obey an equation of state in which $\sigma/|1-T/T_C|^{\beta}$ is uniquely related to $H/|1-T/T_C|^{\beta+\gamma}$, where β and γ are the critical exponents for the temperature dependence of the spontaneous moment and initial susceptibility, respectively.

An important new development concerning the nature of continuous phase transitions has arisen from the prediction of a particularly simple form for the equation of state in the critical region.[1,2] It has been proposed[3] that the magnetization-field-temperature (σ-H-T) behavior of a ferromagnet near its Curie point T_C (or the analogous critical behavior of a gas-liquid system) may conform to a unique relationship between the normalized quantities

$$m \equiv \sigma/|1-T/T_C|^{\beta}$$

and

$$h \equiv H/|1-T/T_C|^{\beta+\gamma},$$

which for later convenience we write as

$$h/m = K_+(m) \text{ for } T \geq T_C$$

and

$$h/m = K_-(m) \text{ for } T \leq T_C. \quad (1)$$

The exponents β and γ are those describing the temperature dependence of the spontaneous magnetization just below T_C,

$$\sigma_s \propto |T-T_C|^{\beta}, \quad (2)$$

and that of the initial susceptibility just above T_C,

$$\chi_0 \propto |T-T_C|^{-\gamma}. \quad (3)$$

Since Eq. (1) must merge at $T = T_C$ with the expression for the critical isotherm,

$$\sigma \propto H^{1/\delta}, \quad (4)$$

it follows that in the limit of large m,

$$K_{\pm}(m) \propto m^{\delta-1}, \quad (5)$$

and that

$$\delta = 1+\gamma/\beta. \quad (6)$$

In the first experimental confirmation of this equation of state, Kouvel and Rodbell[4] showed that above T_C their $\sigma(H, T)$ data on CrO_2 and similar data by Weiss and Forrer[5] on nickel were both closely described by Eq. (1), even though the critical exponents for these two materials are quantitatively very different. They also showed that the limiting behavior for small m was given by

$$K_+(m) = A_0 + A_1 m^2, \quad (7)$$

which has the appropriate form if χ_0 and related higher-order properties are to behave properly near T_C.[6] Subsequently, Arrott and Noakes[7] reported that their own data on a nickel crystal and the forementioned Weiss and Forrer data both gave good agreement above and below T_C with

$$K_{\pm}(m) = (\pm B_0 + B_1 m^{(\delta-1)/\gamma})^{\gamma}, \quad (8)$$

except when m is small. For small m and $T > T_C$, Eq. (8) becomes

$$K_+(m) \approx B_0^{\gamma} + \gamma B_0^{\gamma-1} B_1 m^{(\delta-1)/\gamma}, \quad (9)$$

which disagrees with Eq. (7) unless it happens (as it does not experimentally for nickel) that $(\delta-1)/\gamma = 2$. With regard to fluids, available p-V-T information on several systems was recently analyzed by Green, Vicentini-Missoni, and Levelt Sengers[8] and found to be consistent above and below the critical point with the analogs of Eqs. (1) and (5); the only comparison with the analog of Eq. (7) showed effectively that A_0 is a constant.

The purpose of this Letter is to present new $\sigma(H, T)$ data for nickel which give very close agreement with Eq. (1) for a single set of exponents above and below T_C. Moreover, this agreement holds over a wide range of m, and the asymptotic forms of $K_{\pm}(m)$ are determined with considerable accuracy.

Our nickel specimen was 99.99% chemically pure, polycrystalline (the magnetic anisotropy

Table I. Critical-point parameters predicted from theory and those for nickel.

	Molecular field model	Ising model	Nickel
β	0.500	0.312	0.378 ± 0.004
γ	1.00	1.25	1.34 ± 0.01
δ	3.00	5.0 [b]	4.58 ± 0.05
m_0/σ_0	1.732 [a]	1.487 [c]	1.422
$\mu_0 h_0/kT_C$	1.732 [a]	1.524 [c]	1.037

[a] For $S = \frac{1}{2}$.

[b] Given in Ref. 13, but a value closer to 5.2 is perhaps more accurate [see M. E. Fisher, Rept. Progr. Phys. 20, 615 (1967)].

[c] For fcc structure (Ref. 13).

near T_C being extremely small), and approximately prolate-spheroidal in shape (for uniform demagnetization). Its magnetization was measured at 2.5°K intervals over the temperature range 615-640°K in fields of 0.5 to 25 kOe. The results plotted as isotherms of σ^2 vs H/σ, with H corrected for demagnetization, gave smooth curves whose intercepts with the σ^2 axis (for $T < T_C$) and the H/σ axis (for $T > T_C$) were reliable measures of $\sigma_S{}^2$ and $\chi_0{}^{-1}$, respectively. The curve of $\chi_0{}^{-1}$ vs T thus derived was analyzed according to a previously prescribed method[9] for determining first T_C and then γ, by which we obtained $T_C = 627.4$°K and the γ value (for $T \to T_C{}^+$) shown in Table I. Using this value for T_C, we then tested our $\sigma_S(T < T_C)$ results against Eq. (2) and our interpolated $\sigma(H, T_C)$ data against Eq. (4) and got excellent agreement with β and δ having the values listed in Table I.

Our values for β, γ, and δ are fairly consistent with earlier results for nickel,[9-11] especially with those reported most recently by Arrott and Noakes.[7] Furthermore, within the narrow error limits indicated in Table I, they numerically obey Eq. (6). We therefore proceeded to use these exponent values in transforming the experimental points for each of our σ^2 vs H/σ isotherms into the corresponding points of m^2 vs h/m, as defined above. The latter points are shown plotted in Fig. 1, where they clearly fall on two universal curves, one for $T < T_C$ and the other for $T > T_C$, thus describing the two branches of Eq. (1). Close examination reveals that both these curves start with finite slopes, as depicted by the dashed lines. Hence, for $T < T_C$ in the limit of small h/m,

$$K_-(m) \propto m^2 - m_0{}^2, \tag{10}$$

with $m_0 \equiv (m)_{h/m \to 0} = 83.3$ emu/g; for $T > T_C$ the limiting behavior for small m is given by Eq. (7) with $A_0 \equiv (h/m)_{m \to 0} = 1.887 \times 10^5$ Oe g/emu so that $h_0 \equiv A_0 m_0 = 15.72 \times 10^6$ Oe. At large m both curves in Fig. 1 gradually approach the dashed curve representing our experimental critical isotherm, as converted by means of Eq. (6) into the form of Eq. (5).

Since Fig. 1 emphasizes the region of large m dominated by the points for T closest to T_C, we have plotted the same results on a log-log basis in Fig. 2 and show that Eq. (1) is very well obeyed over the entire range of the normalized variables. The limiting behavior for large and small m (or h/m) is also clearer from Fig. 2, where

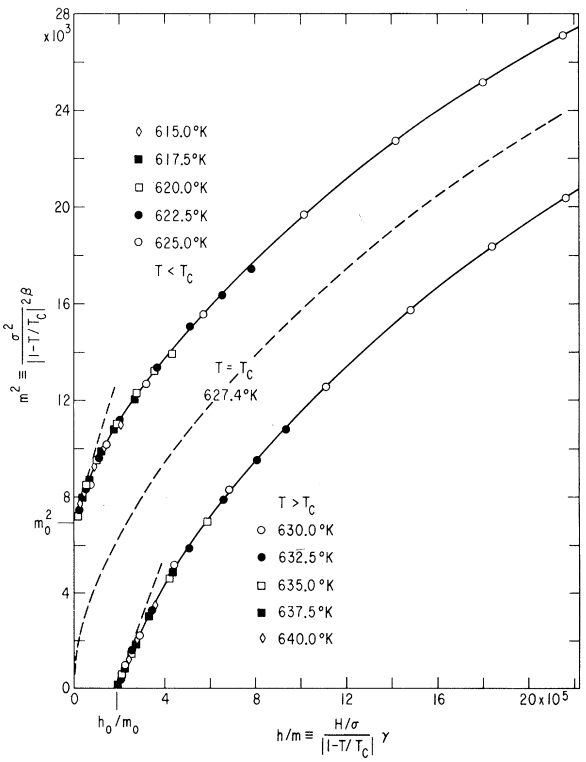

FIG. 1. Normalized isotherms for nickel below and above T_C, where m is in emu/g and H in Oe. Dashed lines and dashed curve indicate limiting behavior at small and large m (or h/m), respectively.

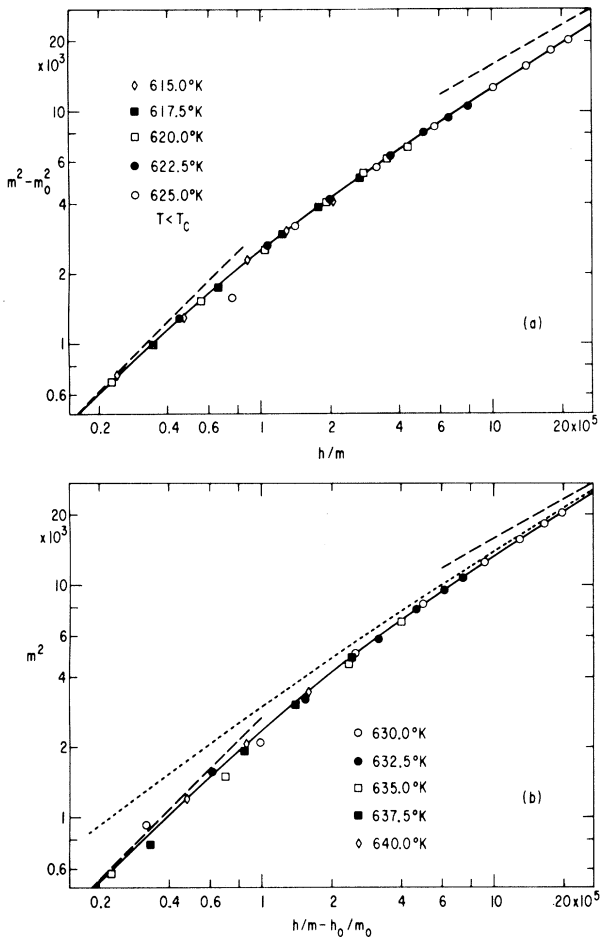

FIG. 2. Log-log plot of normalized isotherms for nickel (a) below T_C and (b) above T_C. Asymptotic behavior indicated by dashed lines. Dotted curve in (b) was calculated from Eq. (8).

the experimental curves approach asymptotically the dashed straight lines representing Eqs. (5), (7), and (10). In an alternative comparison, we have fitted Eq. (8) to our results for $T > T_C$ (by setting $B_0{}^{\gamma}$ equal to our A_0 and evaluating B_1 from our critical isotherm) and obtained the dotted curve in Fig. 2(b); it is obvious that this equation is valid for nickel only at very large values of m.

Finally, in order to compare our above results for m_0 and h_0 with existent theory, we convert them to the dimensionless quantities m_0/σ_0 and $\mu_0 h_0/kT_C$, using $\sigma_0 = 58.6$ emu/g ($\mu_0 = 0.616\mu_B$) for the saturation moment of nickel at 0°K.[12] Their values are listed in Table I together with the corresponding values from the molecular-field model and those recently calculated for the Ising model by Domb.[13] In this context our results appear quite reasonable, considering that both these models predict critical exponents different from the experimental values for nickel (see Table I). Unfortunately, for the Heisenberg model, whose prediction for γ is very close to the measured value for nickel, analogous calculations of m_0 and h_0 (or even of β and δ) have not yet been reported.

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[1]C. Domb and D. L. Hunter, Proc. Phys. Soc. (London) 86, 1147 (1965).

[2]B. Widom, J. Chem. Phys. 43, 3898 (1965).

[3]Equation (1) is essentially in the form proposed in Ref. 1 and, as shown by M. E. Fisher, J. Appl. Phys. 38, 981 (1967), and R. B. Griffiths, Phys. Rev. 158, 176 (1967), it is exactly equivalent to the equation of state proposed in Ref. 2.

[4]J. S. Kouvel and D. S. Rodbell, Phys. Rev. Letters 18, 215 (1967), and J. Appl. Phys. 38, 979 (1967).

[5]P. Weiss and R. Forrer, Ann. Phys. (Paris) 5, 153 (1926).

[6]If $K_+(m)$ is a power series in m^2, all $(\partial^n\sigma/\partial H^n)_{H\to 0}$ that are nonzero (i.e., for n odd) will vary as $|T-T_C|^{-\gamma_n}$ with $\gamma_n = \gamma + (n-1)(\beta+\gamma)$. Such a behavior has been predicted by J. W. Essam and M. E. Fisher, J. Chem. Phys. 38, 802 (1963), and in Ref. 1.

[7]A. Arrott and J. E. Noakes, Phys. Rev. Letters 19, 786 (1967).

[8]M. S. Green, M. Vicentini-Missoni, and J. M. H. Levelt Sengers, Phys. Rev. Letters 18, 1113 (1967).

[9]J. S. Kouvel and M. E. Fisher, Phys. Rev. 136, A1626 (1964).

[10]S. Arajs, J. Appl. Phys. 36, 1136 (1965).

[11]J. E. Noakes and A. Arrott, J. Appl. Phys. 38, 973 (1967).

[12]H. Danan, A. Herr, and A. J. P. Meyer, J. Appl. Phys. 39, 669 (1968).

[13]C. Domb, J. Appl. Phys. 39, 620 (1968). In Domb's notation, m_0/σ_0 and $\mu_0 h_0/kT_C$ are given as $w_0{}^{-1}$ and $b_1 w_0{}^{-1}$, respectively.