

pling $(g_\theta)^2$ alone produces a first-order Jahn-Teller phase transition at 6 °K. When we include the competing biquadratic coupling I_θ we find no phase transition at all down to $T = 0.3$ °K (our numerical calculations only go down to this temperature). However, the elastic constant c_θ does not continue to soften past 9.5 °K because of the bilinear interactions present in DySb. These interactions do not contribute appreciably to the softening of the elastic constant [except for short-range effects, i. e., as corrections to Eq. (4)] but they do cause the system to undergo a first-order phase transition at 9.5 °K.

Finally, by using the values of g_θ and I_θ determined from the elastic data and taking into account the contributions to g_θ that appear below the transition, we have been able to fit¹³ the low-temperature data on the magnetization, susceptibility, specific heat,³ and anisotropic distortion.¹

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¹F. Lévy, Phys. Kondens. Mater. 10, 86 (1969).

²P. M. Levy and H. H. Chen, Phys. Rev. Lett. 27, 1385 (1971); AIP Conf. Proc. 5, 373 (1972).

³E. Bucher, R. J. Birgeneau, J. P. Maita, G. P. Felcher, and T. O. Brun, Phys. Rev. Lett. 28, 746 (1972).

⁴B. Lüthi and M. E. Mullen report that they have obtained similar results on another sample of DySb (private communication).

⁵The samples were prepared at IBM by R. J. Gambino.

These same samples were also used in the neutron scattering work done at Argonne, see Ref. 3.

⁶T. J. Moran and B. Lüthi, Phys. Rev. 187, 710 (1969).

⁷R. J. Elliott, R. T. Harley, W. Hayes, and S. R. P. Smith, Proc. R. Soc. Lond. A 328, 217 (1972).

⁸S. J. Allen, Jr., Phys. Rev. 167, 492 (1968).

⁹E. R. Callen and H. B. Callen, Phys. Rev. 129, 578 (1963); Phys. Rev. 139, A455 (1965).

¹⁰J. Kanamori, J. Appl. Phys. 31, 14S (1960).

¹¹P. M. Levy (unpublished).

¹²G. T. Trammell, Phys. Rev. 131, 932 (1963). We assume a dielectric constant of one.

¹³L. F. Uffer, P. M. Levy, and H. H. Chem. AIP Conf. Proc. (to be published).

Divergence of the Correlation Length along the Critical Isotherm*

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Three plausible postulates (rigorous for Ising systems) are shown to lead to three new inequalities: (i) $(2-\eta)\mu_\phi \geq (\delta-1)/\delta$, (ii) $\mu_\phi \geq 2/\delta(d-2+\eta)$, and (iii) $d\mu_\phi \geq (\delta+1)/\delta$, concerning μ_ϕ , the critical-point exponent characterizing the divergence along the critical isotherm of the correlation length $\xi_\phi(T, H) \equiv [\sum_{\vec{r}} |\vec{r}|^{2\phi} C_2(T, H, \vec{r}) / \sum_{\vec{r}} C_2(T, H, \vec{r})]^{1/2\phi}$. Result (iii) for μ_ϕ is an analog, for the critical isotherm, of the Josephson inequalities. If we make the plausible but unproved assumption that μ_ϕ is independent of ϕ , inequality (i) becomes an equality!

The divergence of a "correlation length" at the critical point ($T = T_c$, $H = 0$) is a hallmark of cooperative phenomena near phase transitions, as it reflects the fact that the correlation function, $C_2(T, H, \vec{r}) \equiv \langle s_0^\alpha s_{\vec{r}}^\alpha \rangle - \langle s_0^\alpha \rangle^2 \equiv \Gamma_2(T, H, \vec{r}) - [\Gamma_1(T, H)]^2$, (1) is becoming extremely long range.¹ We define a family of correlation lengths $\xi_\phi(T, H)$ through the relation²

$$[\xi_\phi(T, H)]^{2\phi} \equiv \frac{\sum_{\vec{r}} |\vec{r}|^{2\phi} C_2(T, H, \vec{r})}{\sum_{\vec{r}} C_2(T, H, \vec{r})}, \quad (2)$$

where $\xi_1(T, H) \equiv \xi(T, H)$ is commonly called the correlation length. The corresponding critical-point exponents are ν'_ϕ , μ_ϕ , and ν_ϕ for the three paths (paths 1-3) defined in Table I; again the conventional exponents ν , ν' , μ correspond to the case $\phi = 1$.

The path-1 and path-3 exponents have been studied considerably more than the path-2 exponents.^{1,2} It is the purpose of this paper to show that one can readily obtain analogs for path 2 of two of the classic exponent inequalities for paths 1 and 3 (relations IIa and II d of Table I). More-

TABLE I. New inequalities proved here for path 2, compared with known inequalities for paths 1 and 3. Here we require $\phi \geq \frac{1}{2}\eta - 1$ (for otherwise η is not defined). Inequalities shown in curly brackets have *not* been proved thus far, to the best of our knowledge. Note that inequalities a and b differ by a reversal of the inequality sign. Note also that if μ_ϕ is independent of ϕ (Postulate D), then inequalities IIa and IIb reduce to scaling-law equalities, while IIc and IID do not.

Path 1 ($T \rightarrow T_c^-, H=0$)	Path 2 ($T=T_c, H \rightarrow 0^+$)	Path 3 ($T \rightarrow T_c^+, H=0$)
Ia $\{\nu'_\phi(2-\eta) \geq \gamma' \text{ ?}\}$	IIa [Eq. (7)]: $\mu_\phi(2-\eta) \geq (\delta-1)/\delta$	IIIa (Ref. 2): $\nu_\phi(2-\eta) \geq \gamma$
Ib (Ref. 5): $\gamma' \geq (2-\eta)\nu'_{\eta/2-1}$	IIb (Ref. 5): $(\delta-1) \geq (2-\eta)\mu_{\eta/2-1}$	IIIb: $\{\gamma \geq (2-\eta)\nu_{\eta/2-1} \text{ ?}\}$
Ic $\{\nu'_\phi \geq 2\beta/(d-2+\eta) \text{ ?}\}$	IIc [Eq. (9)]: $\mu_\phi \geq 2/\delta(d-2+\eta)$	no analogy
Id (Ref. 7): $d\nu'_\phi \geq 2-\alpha'$	IID [Eq.(10)]: $d\mu_\phi \geq (\delta+1)/\delta$	IIId (Ref. 7): $d\nu_\phi \geq 2-\alpha$

over, one can obtain an additional inequality (relation IIc of Table I) for which the analogous inequality has not been proved for paths 1 and 3.

The new results for path 2 require for their proof the following plausible assumptions (which are rigorous for the Ising model^{3,4}):

*Postulate A.*³ For all finite T and positive H , $\Gamma_1(T, H) = M(T, H) = \langle s_0^z \rangle \geq 0$ and $C_2(T, H, \vec{r}) \geq 0$.

*Postulate B.*³ $\Gamma_1(T, H)$ and $\Gamma_2(T, H, \vec{r}) = \langle s_0^z s_{\vec{r}}^z \rangle$ are monotonic increasing functions of H for fixed T and are monotonic decreasing functions of T for fixed H .

*Postulate C.*⁴ $C_2(T, H, \vec{r})$, defined in (1), is monotonic decreasing in H for $T = T_c$.

In addition to the exponent μ_ϕ defined above [$\xi_\phi(T_c, H) \sim H^{-\mu_\phi}$], we introduce an exponent η_ϕ , defined by $X_\phi(T_c, H=0, R) \sim R^{2-\eta_\phi+2\phi}$, to characterize the decay of the correlation function at the critical point; here

$$X_\phi(T, H, R) \equiv \sum_{|\vec{r}| \leq R} |\vec{r}|^{2\phi} C_2(T, H, \vec{r}) . \quad (3)$$

The exponents η_ϕ are independent of ϕ , and we write⁵ $\eta \equiv \eta_\phi$; this relation is valid only for $\phi \geq \frac{1}{2}\eta - 1$, for otherwise η is not defined.

The proof of relation IIa (for path 2) begins with the observation [Postulate C] that $C_2(T_c, 0, \vec{r}) \geq C_2(T_c, H, \vec{r})$; this implies $X_\phi(T_c, 0, R) \geq X_\phi(T_c, H, R)$, and hence

$$X_\phi(T_c, 0, R) \geq [\xi_\phi(T_c, H)]^{2\phi} \bar{\chi}(T_c, H) - \sum_{|\vec{r}| \geq R} |\vec{r}|^{2\phi} C_2(T_c, H, \vec{r}) , \quad (4)$$

where we have used (3), (2), and the fact that the denominator of (2) is the reduced susceptibility $\bar{\chi}(T, H)$. Considering first the case $\phi < 0$, we have

$$\begin{aligned} \sum_{|\vec{r}| \geq R} |\vec{r}|^{2\phi} C_2(T_c, H, \vec{r}) &\leq R^{2\phi} \sum_{|\vec{r}| \geq R} C_2(T_c, H, \vec{r}) \\ &\leq R^{2\phi} \bar{\chi}(T_c, H) , \end{aligned} \quad (5)$$

where the second inequality follows from Postulate A. Substituting (5) into (4), we have

$$X_\phi(T_c, 0, R) \geq [\xi_\phi(T_c, H)]^{2\phi} \bar{\chi}(T_c, H) \times \{1 - [R/\xi_\phi(T_c, H)]^{2\phi}\} . \quad (6)$$

Now R is an arbitrary number; choosing $R = c\xi_\phi(T_c, H)$, with $c > 1$, ensures that the factor represented by the curly brackets in (6) be a positive number. Taking the limit $H \rightarrow 0$ in (6) yields $(2-\eta+2\phi)\mu_\phi \geq 2\phi\mu_\phi + (\delta-1)/\delta$, or

$$(2-\eta)\mu_\phi \geq (\delta-1)/\delta \quad (\text{relation IIa}). \quad (7)$$

To derive (7) for $\phi > 0$, one may repeat the same procedure [cf. Eqs. (38)–(43) of Ref. 2], or one may utilize the fact that $\mu_x \geq \mu_y$ if $x \geq y$ [Lemma I, Ref. 5; note that the validity of this lemma is not restricted to positive x, y].

Relation IIc of Table I is obtained by combining (7) with Eq. (7b) of Ref. 5;

$$\frac{\delta-1}{\delta} \geq \frac{2(2-\eta)}{\delta(d-2+\eta)} + 2\phi \left\{ \frac{2}{\delta(d-2+\eta)} - \mu_\phi \right\} , \quad (8)$$

with the result

$$\mu_\phi \geq \frac{2}{\delta(d-2+\eta)} \quad (\text{relation IIc}); \quad (9)$$

here d denotes the lattice dimensionality.⁶

Inequality (9) may be written in the form $d\mu_\phi \geq 2/\delta + (2-\eta)\mu_\phi$, and combining this with (7) we have

$$d\mu_\phi \geq (\delta+1)/\delta \quad (\text{relation IID}) . \quad (10)$$

Note from Table I that the relations for paths 1 and 3 analogous to (10) are the Josephson⁷ inequalities, $d\nu' \geq 2-\alpha'$ and $d\nu \geq 2-\alpha$. However, Josephson required for his derivation assumptions that have not been proved for any conventional model system and which are sufficiently unobvious that another author^{7(b)} has recently rederived the path-3 Josephson inequality using an altogether different argument (though even the latter derivation requires the assumption $\alpha \geq \alpha'$).

The two-exponent scaling laws,¹ such as

$$d \frac{\delta-1}{\delta+1} = 2-\eta \quad (11)$$

between δ and η , have been questioned by many authors, both because series expansions suggest these laws fail by 1–2% for some three-dimensional lattice models, and because they require for their validity assumptions in addition to that of homogeneity of $C_2(T-T_c, H, \vec{r})$. We next show that the two-exponent scaling law (11) cannot hold unless $B(\phi) \equiv 2/\delta(d-2+\eta) - \mu_\phi$ is identically equal to zero for all values of ϕ in the range $0 \geq \phi \geq \frac{1}{2}\eta - 1$. To see this, we observe that $B(\phi)$ is the

quantity in the curly brackets of (8), and hence the product $2\phi B(\phi)$ is a "correction term" to the Buckingham-Gunton-Stell inequality.⁶ But according to (9), $B(\phi) \leq 0$, so that this correction term is *positive* for all *negative* ϕ .

Note from (8) and (9) that we may obtain lower and upper bounds on μ_ϕ for $0 \geq \phi \geq \frac{1}{2}\eta - 1$,

$$-\frac{1}{2\phi} \left\{ 1 - \frac{d+2-\eta}{\delta(d-2+\eta)} \right\} \geq \mu_\phi - \frac{2}{\delta(d-2+\eta)} \geq 0. \quad (12)$$

For the $d=3$ Ising system, series expansions suggest¹ that $\delta \geq 5$, and $\eta \approx 0.041$, whence (12) implies $0.4 - 0.025/\phi \geq \mu_\phi \geq 0.4$ (this enables us to pin down the numerical value of μ_ϕ to within 10% providing $-\frac{1}{2} \geq \phi \geq \frac{1}{2}\eta - 1$).

Conversely, for those systems (such as the $d=2$ Ising model) for which (11) is satisfied as an equality, $B(\phi)$ must be zero for negative ϕ ; i. e.,⁸

$$\mu_\phi = \frac{2}{\delta(d-2+\eta)} \quad (0 \geq \phi \geq \frac{1}{2}\eta - 1). \quad (13)$$

Note from (12) that μ_ϕ is independent of ϕ over the range indicated!

The last rigorous result,

$$(2-\eta)\mu_{\eta/2-1} = (\delta-1)/\delta, \quad (14)$$

follows from (7) and (8) [cf. relation IIb of Table I]. Since (14) holds for the smallest value of ϕ and μ_ϕ is a monotonic increasing function of ϕ , it follows that if (14) is observed to hold for some particular value of ϕ , $\phi = \phi_0$ (e. g., $\phi_0 = 1$), then (14) must hold for the entire range $\phi_0 \geq \phi \geq \frac{1}{2}\eta - 1$.

In light of the above discussion [especially (13)] it might seem reasonable to make the further postulate that μ_ϕ is independent of ϕ :

Postulate D. $\mu_\phi = \mu$ for all ϕ .

Then (14) implies that

$$(2-\eta)\mu = (\delta-1)/\delta \quad (15)$$

which is a prediction of homogeneity (unlike the equalities obtained by replacing the inequalities by equalities in relations IIc and II d). To the best of our knowledge, this is the first "derivation" of a scaling law (i. e., an exponent relation predicted by the scaling hypothesis) from a clearly defined and plausible set of mathematical assump-

tions (viz., Postulates A-D). In particular, the *only assumption required for (15) that has not been shown to be rigorously true for the Ising model is Postulate D*⁹: to suggest the *possible* validity of *Postulate D*, we have Eq. (13) (and, of course, the heuristic arguments supporting the scaling hypothesis).

The principal new inequalities of this paper are summarized in Table I as relations IIa, IIc, and II d (path 2). Table I was constructed so that the reader can easily see that many of the results proved here for path 2 have not been proved yet for path 1 and/or path 3. Some (perhaps all) of these path-2 results may hold for paths 1 and 3, but we have not yet seen how to prove any of them without making additional assumptions.

One such assumption which, when combined with Postulates A and B, leads to all unproved relations for path 1 (Ia, Ic, and Id) is the following:

Postulate C'. $C_2(T, H=0, \vec{r})$, defined in (1), is monotonic increasing in T for $T \leq T_c$.

Under Postulates A, B, and C' the proof of the results along path 1 is exactly the same as those along path 2. This similarity may be visualized by comparing the contents of the assumptions made in both cases. Postulate A indicates that the spin averages Γ_1 and Γ_2 are always positive, be it along path 1 or 2. Postulate B indicates that Γ_1 and Γ_2 decrease as one approaches the critical point along both paths. Postulate C indicates that the fluctuation $C_2(T, H, \vec{r})$ increases as one approaches the critical point along path 2, and Postulate C' is exactly the analog of Postulate C along path 1. Postulate C' has not been proved (to our knowledge) for the Ising model, unlike Postulates A-C^{3,4}; however, it is a plausible assumption to make, since the fluctuations are "thought" to increase as $T \rightarrow T_c$. This thought is also consistent with the few rigorous results known for the two-dimensional Ising model.¹⁰

The major point of this paper is to emphasize that thorough consideration of path 2 merits the attention of both theoreticians and experimentalists; in particular, numerical experiments are underway in order to determine values of μ_ϕ for system Hamiltonians (Ising, Heisenberg, ...) of direct experimental relevance.

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¹See, e.g., H. E. Stanley, *Introduction to Phase Transitions and Critical Phenomena* (Oxford U. P., London, 1971), and references contained therein.

²M. E. Fisher, *Phys. Rev.* **180**, 594 (1969).

³R. B. Griffiths, *J. Math. Phys.* **8**, 478 (1967); *J. Math. Phys.* **8**, 484 (1967); D. G. Kelly and S. Sherman, *J. Math. Phys.* **9**, 466 (1968); R. B. Griffiths, *J. Math. Phys.* **10**, 1559 (1969).

⁴R. B. Griffiths, C. A. Hurst, and S. Sherman [*J. Math.*

Phys. **11**, 790 (1970)] have proved Postulate C for all T .

⁵L. L. Liu, R. I. Joseph, and H. E. Stanley, *Phys. Rev. B* **6**, 1963 (1972).

⁶Note that the Buckingham-Gunton-Stell inequality $d(\delta-1)/(\delta+1) \geq 2-\eta$ is a special case of (8) with $\phi=0$.

⁷(a) B. D. Josephson, *Proc. Phys. Soc. Lond.* **92**, 269 (1967); *Proc. Phys. Soc. Lond.* **92**, 276 (1967). We have observed that (10) also follows from applying the Josephson-type argument to path 2, but, as indicated in the text, this approach is far less appealing than that of using Postulates A, B, and C. (b) G. Stell, *Phys. Rev. B* **6**, 4207 (1972).

⁸Equation (13) also follows directly from (12) since the quantity in curly brackets is zero for those systems satisfying (11).

⁹Note that Postulates A–D *do not* predict that relations II c and II d of Table I become equalities, despite the fact that II c and II d *do* become equalities in two-exponent scaling theory. This is not inconsistent with the current idea that homogeneity

(and the consequent three-exponent relations) may be generally valid, but that the *additional assumption(s) required to obtain the two-exponent scaling relations are not generally valid*. See, e.g., A. Hankey and H. E. Stanley, Phys. Rev. B 6, 3515 (1972), and references therein.

¹⁰R. Dekeyser and J. Rogiers, Physica (Utr.) 59, 23 (1972).

Resistivity Studies of Antiferromagnetic Mn-Rich Alloys Containing up to 2.5-at.% V, Cr, Fe, and Ru

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In an attempt to better understand the nature of the complex magnetic interaction in antiferromagnetic α -Mn, we have undertaken an experimental determination of the Néel temperatures (T_N) of manganese-rich Mn-V, Mn-Cr, Mn-Fe, and Mn-Ru alloys. The electrical resistivities of Mn samples containing up to approximately 2.5 at.% of V, Cr, Fe, and Ru have been studied from 4.2 K to room temperature. Anomalies in the temperature dependence of the alloy resistivities at the onset of antiferromagnetic ordering reveal that dissolving V or Cr into Mn depresses T_N , while dissolving Fe or Ru into Mn raises the Néel temperature of the alloy. The data reveal that the introduction of impurity atoms into α -Mn does not affect the antiferromagnetic coupling by a simple dilution process. T_N exhibits a striking dependence on both the magnitude and sign of the excess electron concentration in the alloys.

INTRODUCTION

α -Mn has a cubic structure with lattice constant 8.9135 Å and contains 58 atoms per unit cell. Bradley and Thewlis¹ showed that the structure contained four crystallographically nonequivalent sites. The basis of the whole arrangement is a simple body-centered-cubic lattice, with each lattice point being associated with a cluster of 29 atoms. Around each type-I atom is an octahedron of type-IV atoms, the opposite faces of the octahedron being of different sizes so that the symmetry is tetrahedral. The four type-II atoms are somewhat further from the center of the group and are arranged tetrahedrally about the center. The twelve outer-most type-III atoms comprise a polyhedron having cubic and octahedral faces. The whole cluster has symmetry which is tetrahedral, as is that of the crystal as a whole.

α -Mn is in many respects analogous to an intermetallic compound. In fact, an intermediate phase generally called the χ phase, has a structure isomorphous with α -Mn. This χ phase has been identified in several binary^{2,3} and ternary alloys.²⁻⁵ Two factors would appear to be in operation in stabilizing both α -Mn and the χ phase. They are electronic structure and atomic sizes.

The coordination numbers (CN) for the α -Mn sites are site-I, CN 16; site II, CN 16; site III, CN 13; and site IV, CN 12.³ There is a striking difference between interatomic distances within the Mn structure, as well as coordination numbers

associated with the various sites. The interatomic distances vary from 2.21 to 2.96 Å³ and the coordination numbers vary from the compact icosahedral with CN 12, to sites with CN 16 which occupy considerably more volume. From consideration of space filling, it would appear that the Mn atoms in sites I and II would tend to have larger electronic radii in order to fill the relatively larger volume of the CN 16 sites. Smaller atoms, on the other hand, would occupy the sites III and IV, since there is a smaller volume associated with the CN 13 and CN 12 sites. From size consideration, it might be expected that Mn exists in different electronic states.

Several neutron-diffraction investigations of the magnetic structure of α -Mn have been done.⁶⁻⁹ These studies have shown that each of the four nonequivalent atom sites have different magnetic moments and established the existence of an antiferromagnetic state in α -Mn below a temperature of approximately 95 K.

From the most recent neutron-diffraction study, in which a noncollinear-spin model was assumed, moments of 1.9 μ_B , 1.7 μ_B , 0.6 μ_B , and 0.2 μ_B were obtained for sites I, II, III, and IV, respectively. This model supported a localized magnetic moment on each of the α -Mn atoms, as opposed to a spin-density wave.

From consideration of atom sizes, predictions can be made with regards to the effects of alloying. One may expect atoms of relatively smaller sizes, such as Fe and Cr, to preferentially occupy the