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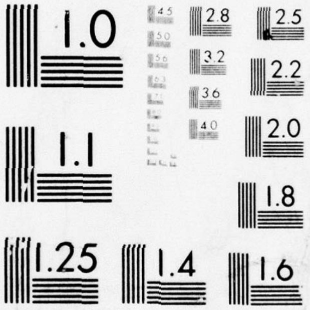

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FINAL REPORT

GRANT NO. 72-2197 ⑮ AF-AFOSR-2197-72

AIR FORCE OFFICE OF SCIENTIFIC RESEARCH

Title of Project

⑥ EQUATION OF STATE AND OTHER PROPERTIES OF REALISTIC MATERIALS IN THE VICINITY OF THEIR CRITICAL POINTS.

Grantee

Massachusetts Institute of Technology

⑪ 31 Dec 76

⑫ 28p.

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Period of Award: Final rept.

1 January 1972 – 31 December 1976,

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⑩ H.Eugene Stanley, Principal Investigator
Hermann von Helmholtz Associate Professor

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REPORT DOCUMENTATION PAGE

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1. REPORT NUMBER AFOSR-TR- 77-0953	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) EQUATION OF STATE AND OTHER PROPERTIES OF REALISTIC MATERIALS IN THE VICINITY OF THEIR CRITICAL POINTS		5. TYPE OF REPORT & PERIOD COVERED Scientific Final 6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) H Eugene Stanley		8. CONTRACT OR GRANT NUMBER(s) AFOSR 72-2197
9. PERFORMING ORGANIZATION NAME AND ADDRESS Department of Physics Massachusetts Institute of Technology Cambridge MA 02139		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 9767-04 61102F
11. CONTROLLING OFFICE NAME AND ADDRESS AFOSR/NP Bolling AFB, Bldg.#410 Wash DC 20332		12. REPORT DATE 13. NUMBER OF PAGES 26
14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office)		15. SECURITY CLASS. (of this report) Unclassified 15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Engineers of all sorts, from steam engines to those used in nuclear power plants, require a knowledge of a) the precise location of phase transition lines, b) "exactly" what happens when a material (such as steam) is moved across a given phase transition line. This information is essential from the engineering point of view because only with this information can one hope to have a truly efficient engine design. Phase transitions have become an extremely active field of research because of this need. Engineers, chemists, physicists, metallurgists, and mathematicians have cooperated in a multidisciplinary → next page

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effort to learn new information on this subject. These efforts can be broadly categorized as follows: a) studies focussed on critical point exponents describing the behavior of a single function as a single variable is changed; b) studies focussed on equation of state describing the behavior of a function when all thermodynamic variables are changed. The focus of this contract has been category (b), equation of state. A simple example of an equation of state is provided by a simple magnet in the vicinity of the Curie temperature Tc.

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FINAL REPORT

Engines of all sorts, from steam engines to those used in nuclear power plants, require a knowledge of

a) the precise location of phase transition lines

b) "exactly" what happens when a material (such as steam) is moved across a given phase transition line.

This information is essential from an engineering point of view because only with this information can one hope to have a truly efficient engine design.

Phase transitions have become an extremely active field of research because of this need. Engineers, chemists, physicists, metallurgists, and mathematicians have cooperated in a multidisciplinary effort to learn new information on this subject. These efforts can be broadly categorized as follows:

a) studies focussed on critical point exponents describing the behavior of a single function as a single variable is changed.

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The focus of this contract has been category (b), equation of state. A simple example of an equation of state is provided by a simple magnet in the vicinity of the Curie temperature T_c.

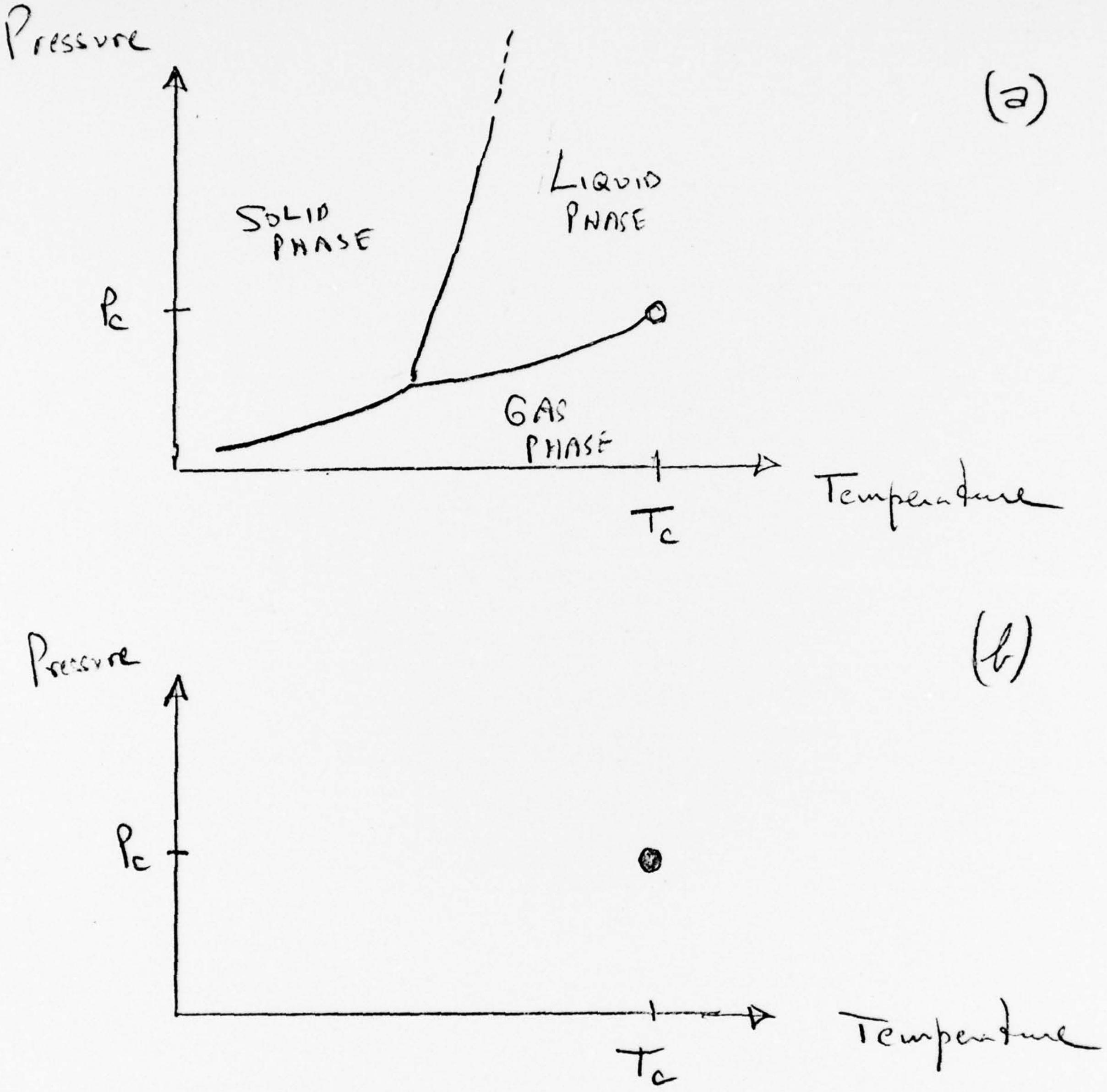

FIGURE 1. PHASE TRANSITIONS & CRITICAL PHENOMENA.

a) Phase transitions occur at all three solid lines separating different phases

b) Critical phenomena occur at the single point (P_c, T_c) at which the first-order phase transition line separating gas and liquid phases terminates.

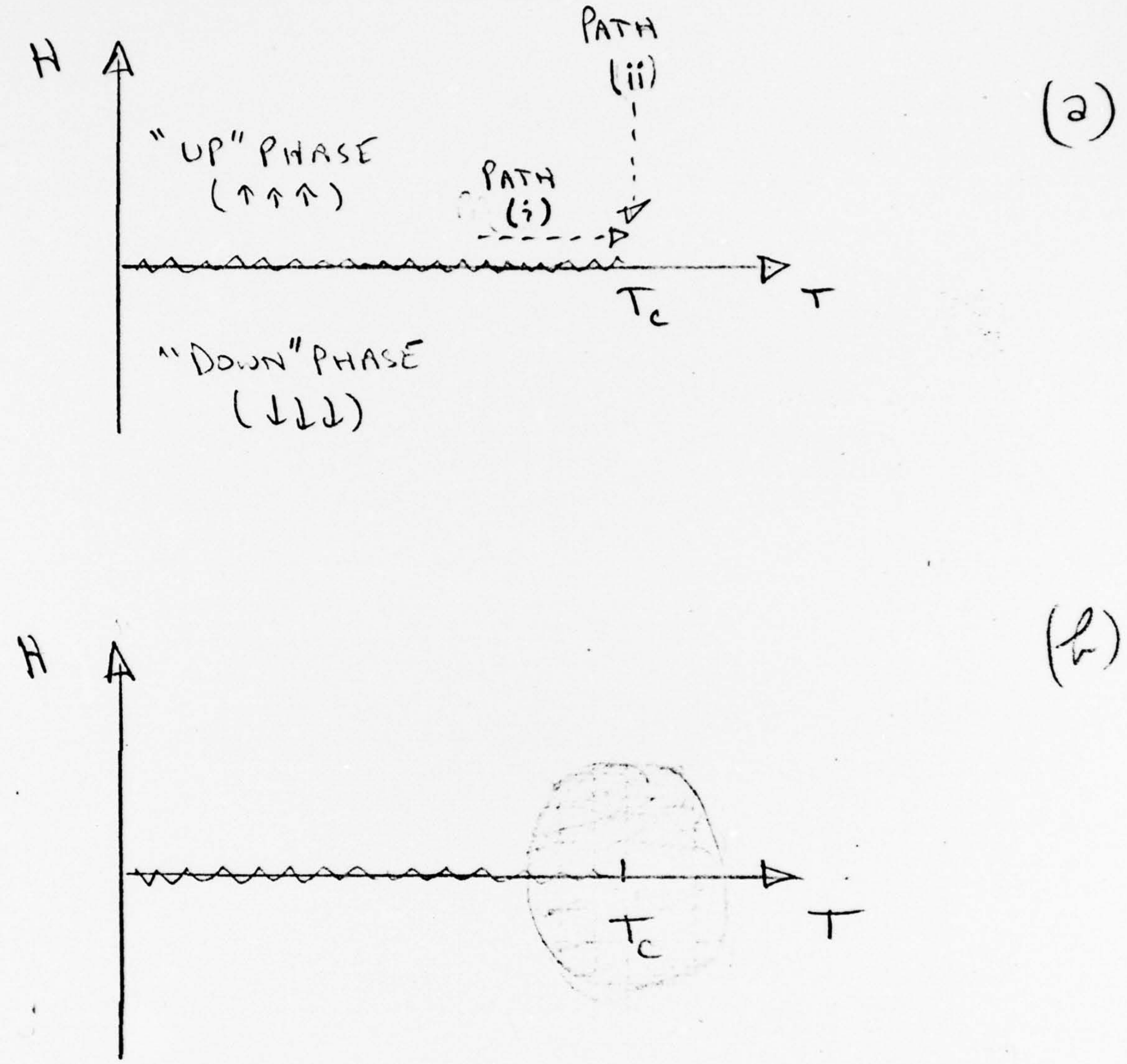

FIGURE 2. DIFFERENCE BETWEEN CRITICAL POINT EXPONENTS AND THE EQUATION OF STATE OF A MAGNET

a) Phase diagram of a magnet, showing information learned by critical point exponents along 2 paths

b) By contrast, the equation of state provides information in the entire phase diagram near the critical point ($H=0, T=T_c$).

The phase diagram of the simple magnet is provided in Fig.2. There is a line of first order transitions which terminates in a critical point at $T=T_c$, $H=0$ where H is the applied magnetic field. Studies of category (a) concern the determination of critical exponents along paths (i) and (ii) of Fig.2a. For example, if the function in question is the spontaneous magnetization, $M = M(H,T)$, then along path (i)

$$M = M(H=0,T) \sim t^{\beta} \qquad (1a)$$

while along path (ii),

$$M = M(H,T=T_c) \sim H^{1/\delta} \qquad (1b)$$

thereby defining the two critical exponents β and δ. Here we have used the notation $t \equiv (T-T_c)/T_c$ to denote the reduced temperature (i.e., *t=0 at the critical point*).

Studies of category (b) concern the determination of the function in the entire region near the critical point. For our example of the magnetization function $M(H,T)$, this means that we must determine M everywhere in the region shown shaded in Fig.2b.

Clearly studies of category (b) provide more information than studies of category (a), and therefore it is not surprising that they are more difficult to carry out.

The principal difficulty stems from the fact that very close to the critical point the material undergoes fluctuations that are not only characterized by their size but also are characterized by the fact that the fluctuations of all wavelengths are simultaneously present. This means that any approximation procedure of the sort familiar from solid state physics is doomed to failure.

Our research group has participated in the development of an approach to the development of a theory of the equation of state. This approach permits the accurate calculation of of the equation of state for a wide range of materials.

Specifically, it has been found that very near the critical point, the equation of state of a wide range of materials is a "generalized homogeneous function" (GHF). A function of two variables H and t is a GHF if there exist two numbers (called scaling powers) a_H and a_t such that for all positive values of some arbitrary parameter L,

$$f(L^{a_H} H, L^{a_t} t) = L\, f(H,t) \qquad (2)$$

Specifically, the function f(H,t) is the Gibbs potential. Since every thermodynamic function is related by a series of Legendre transformations and partial differentiations to the Gibbs potential, Equation (2) has far-reaching implications. Specifically, the magnetization function M(H,t) is related to the Gibbs potential by the relation

$$M = \left[\partial f(H,t)/\partial H\right] \quad \text{at } t=\text{const} \qquad (3)$$

Therefore from (2) and (3) it follows that M(H,t) is also a GHF; that is,

$$M(L^{a_H} H, L^{a_t} t) = L^{1-a_H} M(H,t) \qquad (4)$$

The physical content of Equation (4) is that the equation of state M(H,t) is constrained rather severely. To see this, consider the equation of state for a typical material. This consists of a series of values of M(T) for various values of H. That is, for each value of H, one must measure M(T). Such data are shown schematically in Fig.3, as a family of curves, one for each value of H.

Now Equation (4) implies that the entire family of curves collapses onto one single curve providing the experimental data are plotted in a "scaled form". To see this, note from (4) that L is an arbitrary number, and therefore can be chosen as

$$L = (1/H)^{1/a_H} \tag{5}$$

Substituting (5) into (4), we obtain

$$\frac{M(H,t)}{H^{(1-a_H)/a_H}} = M(1, \ t / H^{a_t/a_H}) \tag{6}$$

But Equation (6) states that if one plots "scaled magnetization"

$$\tilde{M} \equiv M/ H^{(1-a_H)/a_H}$$

as a function of "scaled temperature"

$$\tilde{t} \equiv t/ H^{a_t/a_H} \tag{7}$$

then one finds that the entire family of curves in Fig.3a "collapse" onto a single curve in Fig.3b.

This single curve is called the "scaled equation of state" or the "scaling function" for short. The experimental verification of the scaled equation of state is rather

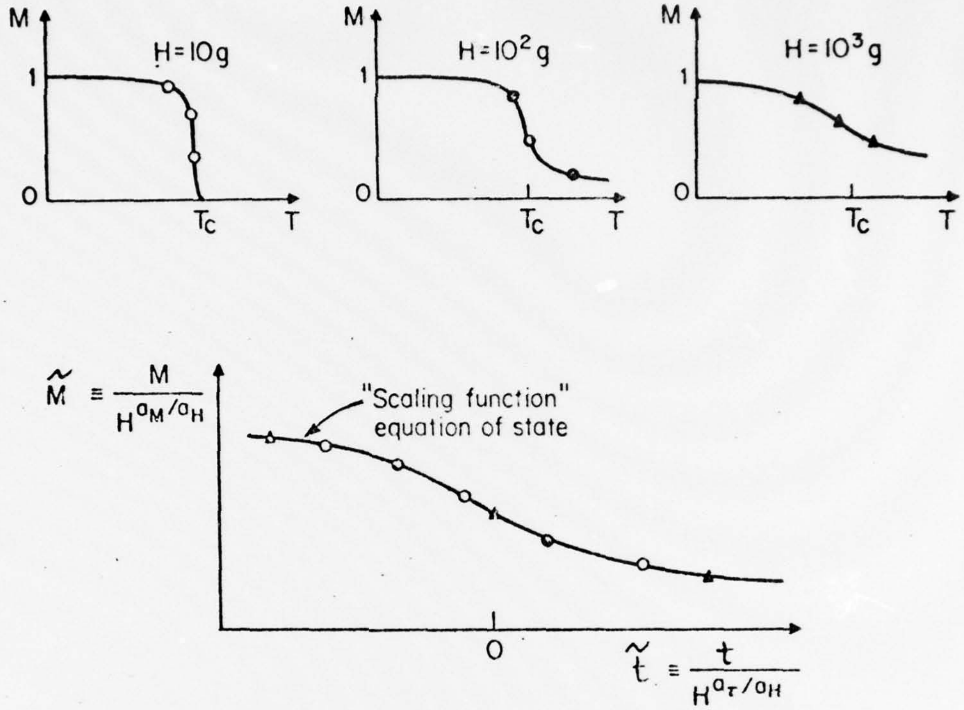

FIGURE 3.

Illustration of "data collapsing" onto a single "scaled equation of state".

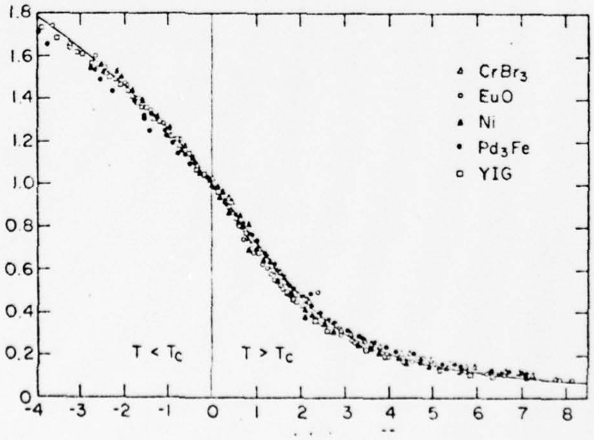

FIGURE 4.

Data collapsing for 5 different materials onto a single scaling function. Solid curve = calculations.

striking, as shown in Figure 4. Here we have plotted experimental data taken on five widely diverse materials The first material, $CrBr_3$,has considerable lattice anisotropy (the coupling strengths in the z lattice direction are about 17 times weaker than those along the x or y directions). The second material, EuO, has signficant second-neighbor interactions, and these are probably of the opposite sign to the nearest-neighbor interactions. The third material, Ni, is probably an itinerant electron ferromagnet, while the fourth material Pd_3Fe is an alloy and the fifth material YIG is actually a ferrimagnet. All five materials display data collapsing, thereby providing experimental confirmation of the scaling equation of state.

From Figure 4, we see that all these data fall on one and only one "scaling function"--i.e., all five materials have the identical scaled equation of state! This fact suggests that while the equation of state is strongly dependent on the details of intermolecular interactions for most regions of the phase diagram, at least near the critical point it appears to be independent of such details for otherwise five such widely diverse materials could hardly obey the identical scaled equation of state!

Such apparent universality has provided impetus for a variety of calculations during the tenure of this grant. Specifically, we have developed the method of high-temperature series expansions in such a fashion that it can be used to provide information on the equation of state of a wide variety of materials. Then we have proceeded to calculate the equation of state and to compare the calculated scaling functions with experimental data. The calculated function is shown in Figure 4 as the solid curve, and we note that the agreement is rather satisfactory--especially considering the fact that there are no free parameters in the theory.

It is perhaps not entirely inappropriate to record the fact that this work has led to invitations to address three major international meetings:

a) the International Conference on Low-Temperature Physics
b) the International Congress on Magnetism, Moscow, USSR
c) the International Enrico Fermo School of Physics, Varenna, Italy

So much for studies of ordinary critical points. It is becoming increasingly appreciated that more complex materials display much more complex critical points. For example, Soviet work reported first in the Russian Journal of Physical Chemistry has stimulated a tremendous increase in our understanding of such complex materials. Specifically, the Soviet work considered materials that consisted of three-component fluids mixed in such proportions that a "tricritical point" can occur.

Subsequent to the early work by Soviet authors, triciritical points have been detected experimentally in an extremely wide range of materials, including not only multicomponent fluid mixtures but also magnetic materials, superconductors, liquid crystals, and so forth.

Our work began with the study of metamagnetic materials, which are characterized by a sudden discontinuous change in magnetization as a function of applied magnetic field. These materials are among the simplest from a theoretical point of view. In partcular, colleagues from the Division of Engineering & Applied Physics of Yale University (W.P.Wolf and co-workers) have made detailed measurements of the equations of state of dysprosium aluminum garnet(DAG), one such metamagnet. A natural theoretical question is the following: "can these data be interpreted in terms of a unifying scaling hypothesis?" To this time, there had been no experimental tests of the scaling hypothesis at *tri*critical points. However in light of the success of the scaled equation of state at an ordinary critical point, we were optimistic and decided to analyze the Yale data on DAG.

We found that the data did indeed collapse onto a single curve, but the properties of this curve were more like those of an ordinary critical point than a tricritical point. Thus our results remained a mystery until Blume and co-workers at Brookhaven National Laboratory realized that the experimental data were obtained under conditions in which the applied field coupled to the order parameter and thereby moved one off the H-T plane. The experiments have recently been repeated with a different orientation or the H field, and the predictions of tricritical scaling confirmed.

The experimental data are plotted in Fig.5, and the general phase diagram of a tricritical point is shown in Fig.6.

In addition to "ordinary" critical points and "tricritical points", we have considered the possibility of even more complex critical points. For example, a tricritical point occurs at the intersection of lines of ordinary critical points. This leads to the question "What sort of critical point would occur at the intersection of lines of tricritical points?"

To answer this question, we have developed a classification of complex critical points that is fairly general (though probably not sufficiently general to handle the most complex materials in nature). Specifically, we have introduced the concept of the "order" of a critical point. Ordinary critical points are of order 2, while tricritical points are of order 3 and the point of intersection of critical lines is of order 4. A metamagnet in the presence of both a direct and a staggered magnetic field can be shown to exhibit a critical point of order 4, and we made calculations of critical exponents for such a system.

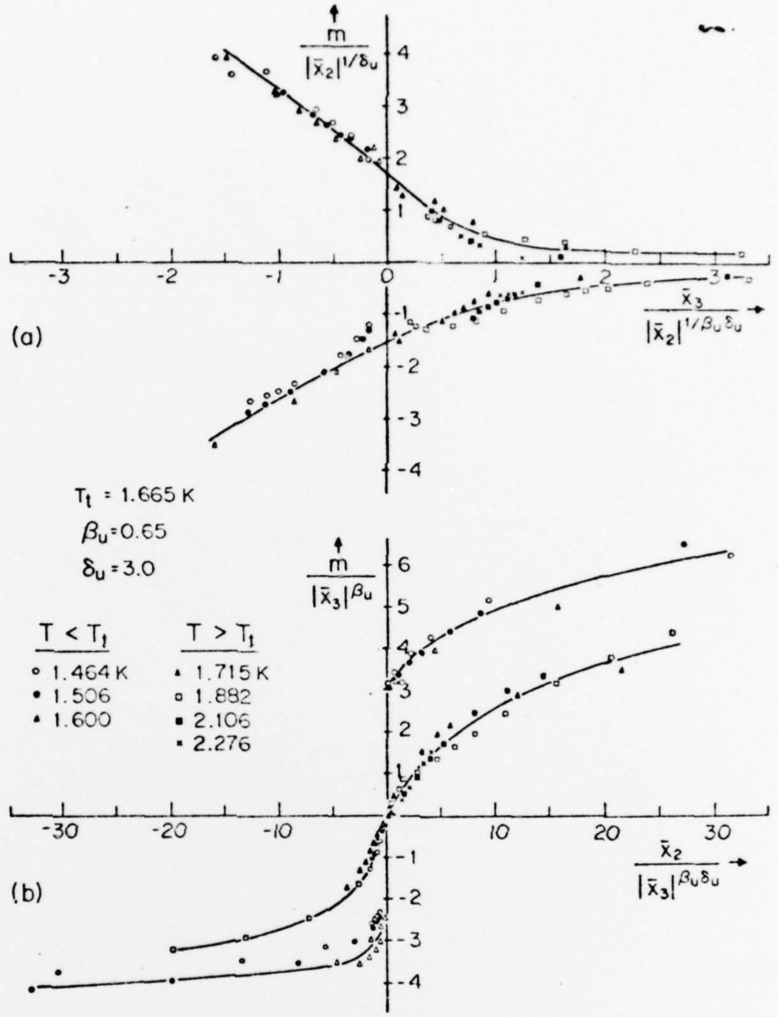

FIGURE 5.

Experimental data of the Yale group on the metamagnetic material dysprosium aluminum garnet (DAG). The data collapse onto a single curve, the scaled equation of state. The data analysis was carried out under the auspices of this grant.

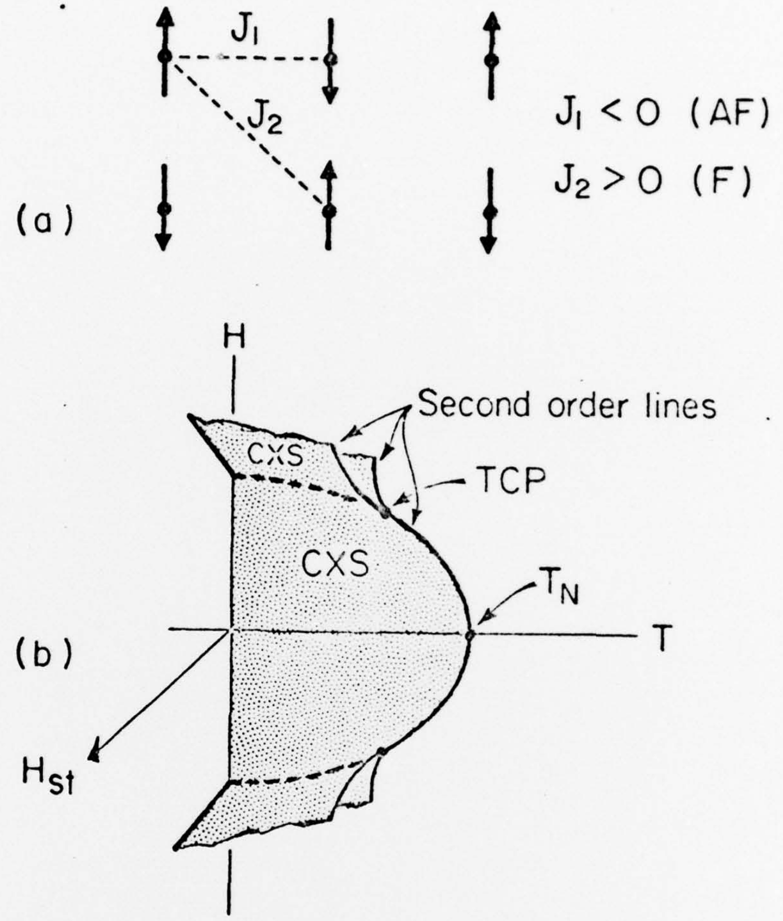

FIGURE 6.

Phase diagram for a typical model metamagnet. Note that three different second order lines of "ordinary" critical points intersect at a "tricritical point" (TCP).

A final class of "realistic materials" will be discussed. These are materials which in one sense or another can be considered to have an effective dimensionality less than 3. Such materials are likely to become extremely important in engineering applications. For example, graphite is in many senses effectively 2-dimensional, and this near 2-dimensionality results in many of its unusual mechanical properties. Similarly, the search for high-temperature superconductors has focussed in part upon materials that are quasi-2-dimensional.

Therefore we have focussed our attention upon a class of materials in which the interactions between spins in one lattice direction are R times those in the remaining 2 directions. For R very small, the system is quasi-2-dimensional, while for R very large the system is quasi-1-dimensional.

Experimental work on materials with R very small has suggested the possible occurrence of a "Stanley-Kaplan phase transition" to a novel low-temperature phase in which there is an infinite susceptibility but no long-range order.

We have developed a set of rigorous relations that relate the measured properties of the full three-dimensional system (i.e., $R \neq 0$) to the two-dimensional system ($R=0$). Similarly, we have been able to predict the location of the crossover from effectively two-dimensional behavior far above the critical temperature to a limiting form of three-dimensional behavior very close to the critical temperature. This work has led to a joint collaboration with L.J.deJongh at the Kamerlingh Onnes Laboratorium, Leiden. deJongh carried out experiments that verified the predictions of the crossover theory, and the results were published jointly with the Principal Investigator in a recent issue of Physical Review Letters.

In summary, then, we have studied a wide variety of materials that are relatively complex compared with the idealized systems that are usually "first" studied by theorists. These systems have included materials such as DAG that display higher order critical points, and materials that display quasi-1-dimensional or quasi-2-dimensional order. It has been found that these materials have an equation of state that obeys a remarkably simple form when expressed in terms of the proper variables.

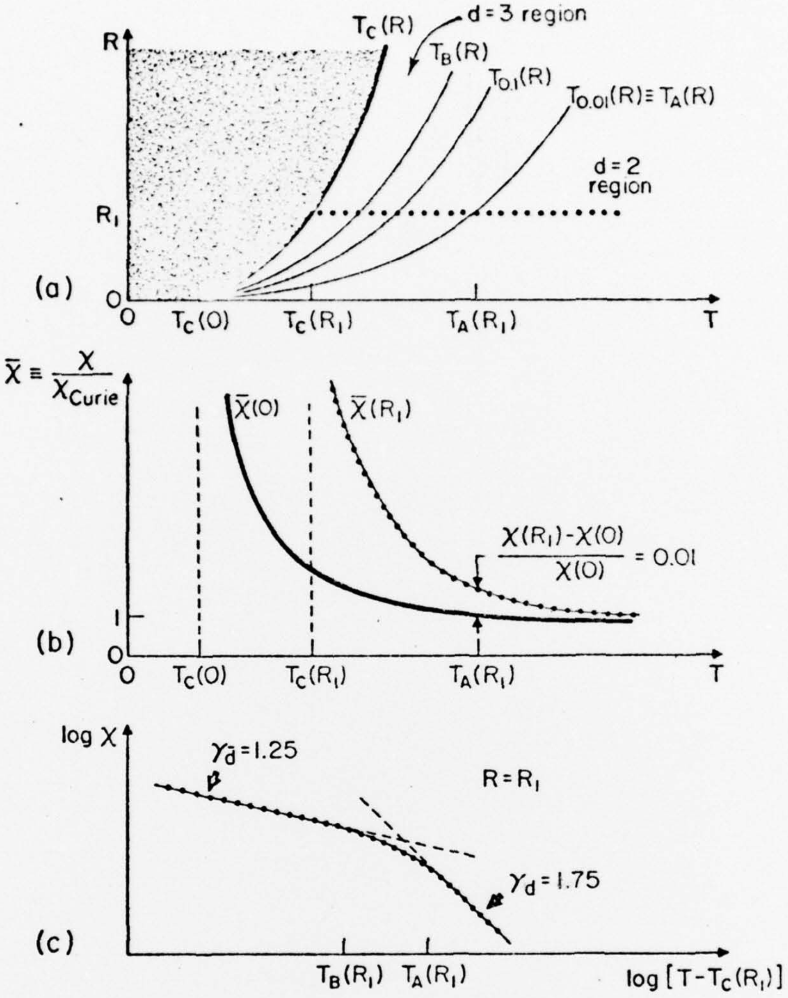

FIGURE 7.

Analysis of crossover phenomena in quasi-2-dimensional magnetic materials. A system changes its critical properties as the critical point is approached arbitrarily closely, in accordance with certain rigorous results of Liu and Stanley (1973) and confirmed experimentally by deJongh and Stanley (1976).

INVITED TALKS BASED ON RESEARCH SUPPORTED BY THE SPONSOR:

1. Invited speaker (with S. Milosevic, D. Karo, and R. Krasnow), Thirteenth International Conference on Low-Temperature Physics ("LT-13"). Boulder, Colorado, August 1972.

2. Principal Invited Speaker (6 intro. lectures), Enrico Fermi Summer School on Local Phenomena near Phase Transitions, Varenna, Italy, July 1973.

3. Invited speaker (with L. Liu), International Workshop on Physics in Less than Three Dimensions, July 1973.

4. Invited speaker, International workshop on Applications of Statistical Mechanics to Cooperative Phenomena, Dartmouth College, August 1973.

5. Invited speaker (with S. Milosevic), International Congress on Magnetism, Moscow, U.S.S.R., August 1973.

6. Invited speaker, Conference on Cooperative Phenomena, Portoroz, Yugoslavia, June 1974.

7. Invited speaker, Latin American School of Physics, Mexico City, July 1974.

8. Invited speaker, AMERICAN PHYSICAL SOCIETY, May 1975 (Kent, Ohio).

9. Invited speaker, NOZAWA SUMMER SCHOOL IN SOLID STATE PHYSICS, NOzawa, Nagano Pref., Japan, July 1975.

10. Invited speaker, INTL. CONF. ON LOW-LYING LATTICE VIBRATIONAL MODES AND THEIR RELATIONSHIP TO SUPERCONDUCTIVITY AND FERROELECTRICITY, San Juan, P.R., 1-5 Dec. 1975.

CONFERENCES AND SUMMER SCHOOLS DIRECTED:

1. International Organizing Committee (with P.W. Anderson, R. Blinc, H.Z. Cummins, J.A. Krumhansl, B.T. Matthias, J.R. Schrieffer, H.G. Smith, and G. Shirane), INTERNATIONAL CONFERENCE ON LOW-LYING LATTICE VIBRATIONAL MODES AND THEIR RELATIONSHIP TO SUPERCONDUCTIVITY AND FERROELECTRICITY, 1-5 December 1975. San Juam, P.R.

2. Director, Summer Program on Phase Transitions and Critical Phenomena, M.I.T., Cambridge, MA, July 1970, June 1971, June, 1972, June 1973, June 1974 and June 1975.

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Translations:

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Translated by S.V. Vonsovsky
(MIR, Moscow, 1973)

Japanese:

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(Tokyo-Tosho, Tokyo, 1974)

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