ple model that has served to explain previous experimental results is inadequate to deal with the latest findings. The  $\vec{k} \cdot \vec{p}$  perturbation method is probably appropriate because a relatively small portion of the zone needs to be considered. This theoretical approach is now being pursued for the purpose of calculating the Landau levels and the selection rules. At this time it is possible to say with reasonable confidence that transitions from impurity levels<sup>11</sup> are not present in these data because not only do we find the main features to scale with frequency but also the necessary carrier freeze-out phenomenon has never been observed in tellurium either at low temperature or high magnetic field.<sup>7</sup> Further experiments are to be attempted using linear and circular polarized radiation to reduce complications that may be introduced by birefringence.

We are indebted to Lawrence G. Rubin for his valuable experimental advice. We are pleased to acknowledge the able assistance of R. E. Newcomb and F. Tambini during the design, construction, and operation of the apparatus. We wish to thank H. J. Engelmann, U. Günther, and R. Drope for their care and skill in the preparation of the specimens. The travel grant for one of us (G.L.) given by the Deutsche Forschungsgemeinschaft is gratefully acknowledged. We are grateful to H. A. Gebbie for his encouragement of submillimeter research. \*Supported by the U. S. Air Force Office of Scientific Research.

†On leave from the National Physical Laboratory, Teddington, England.

‡Also Physics Department, Massachusetts Institute of Technology, Cambridge, Mass. 02139.

<sup>1</sup>J. H. Mendum and R. N. Dexter, Bull. Am. Phys. Soc. <u>9</u>, 632 (1964).

<sup>2</sup>J. C. Picard and D. L. Carter, J. Phys. Soc. Japan Suppl. <u>21</u>, 202 (1966).

 ${}^{3}$ P. L. Radoff and R. N. Dexter, Bull. Am. Phys. Soc. <u>14</u>, 330 (1969).  ${}^{4}$ E. Braun and G. Landwehr, J. Phys. Soc. Japan

<sup>4</sup>E. Braun and G. Landwehr, J. Phys. Soc. Japan Suppl. <u>21</u>, 380 (1966); C. Guthmann and J. M. Thuillier, Solid State Commun. <u>6</u>, 835 (1968); M. S. Bresler, I. I. Farbstein, D. V. Mashovets, Yu. V. Kosichkin, and V. G. Veselago, Phys. Letters 29A, 23 (1969).

<sup>5</sup>K. J. Button, H. A. Gebbie, and B. Lax, IEEE J. Quantum Electron. 2, 202 (1966); C. C. Bradley, K. J. Button, B. Lax, and L. G. Rubin, IEEE J. Quantum Electron. 4, 733 (1968).

<sup>6</sup>K. J. Button, B. Lax, and C. C. Bradley, Phys. Rev. Letters <u>21</u>, 350 (1968); K. J. Button, A. Brecher,

B. Lax, and C. C. Bradley, to be published.

<sup>7</sup>P. Grosse, Z. Physik <u>193</u>, 318 (1966).

<sup>8</sup>M. Picard and M. Hulin, Phys. Status Solidi <u>23</u>, 563 (1967).

<sup>9</sup>P. Grosse and K. Winzer, Phys. Status Solidi <u>26</u>, 139 (1968).

 $^{10}$ K. J. Button, G. Landwehr, C. C. Bradley, B. Lax, and D. Cohn, to be published.

<sup>11</sup>Y. Couder, Phys. Rev. Letters <u>22</u>, 890 (1969).

## NONANALYTIC BEHAVIOR ABOVE THE CRITICAL POINT IN A RANDOM ISING FERROMAGNET\*

## Robert B. Griffiths<sup>†</sup>

Institute for Theoretical Physics, State University of New York at Stony Brook, Stony Brook, New York 11790 (Received 3 June 1969)

It is shown that in a class of randomly diluted Ising ferromagnets the magnetization fails to be an analytic function of the field H at H=0 for a range of temperatures above that at which spontaneous magnetization first appears.

It is commonly assumed<sup>1</sup> that the critical point of a simple ferromagnet represents a complete termination of the first-order phase transition indicated by the presence of spontaneous magnetization, and above the critical temperature  $T_c$ where this spontaneous magnetization disappears, the free energy is an analytic function of temperature T and magnetic field H. In other words, for  $T > T_c$  there is no "higher-order" transition representing a continuation of the first-order transition for  $T < T_c$ . We shall show that in the case of certain random Ising ferromagnets this assumption is demonstrably false, and the temperature  $T_c$ , where nonanalyticity begins as T decreases definitely exceeds  $T_c$ .

The random ferromagnets of interest are obtained by starting with an ordinary or <u>regular</u> Ising model with spins located on the vertices of a regular two- or three-dimensional lattice, with nearest-neighbor exchange interactions. In the corresponding random ferromagnet<sup>2</sup> only a fraction p of the lattice sites are occupied with Ising spins, the rest remaining vacant, and exchange interactions exist only between spins on neighboring pairs of occupied sites.<sup>3</sup> The probability p of occupancy of a given site is independent of H, T,



FIG. 1. Probable behavior of  $T_c$  (solid curve) and  $T_c^*$  (dashed curve) in a randomly dilute Ising ferromagnet.

and the occupancy of other sites; this last condition distinguishes the model from the apparently similar but mathematically quite different model of Syozi<sup>4</sup> in which an average occupation is assured through the imposition of a suitable "chemical potential."

It is fairly well established that  $T_c$  for the random ferromagnet varies with p at least qualitatively in the fashion indicated in Fig. 1. Evidence in favor of this behavior is (1) the model of Syozi referred to earlier which might be expected to show qualitatively the same dependence of  $T_c$  on p; (2) studies based on series expansions<sup>5</sup>; (3) a rigorous upper bound<sup>6</sup> obtained by applying Fisher's procedure<sup>7</sup> to the random case; (4) a rather inefficient rigorous lower bound<sup>2</sup>; and (5) a proof<sup>6</sup> of the "intuitively obvious" result that no spontaneous magnetization can occur at a finite temperature with p less than the critical percolation probability  $p_o$  at which an "infinite cluster" first appears as p increases.

However, we shall show that for any concentration p < 1 the magnetization M is not an analytic function of H at H = 0 at any temperature below  $T_c(1)$ , the critical temperature of the regular Ising model (p = 1); that is, in the region below the dashed line in Fig. 1. The argument is most easily explained for  $p < p_o$  when the magnetization Mper lattice site in the thermodynamic limit has the form<sup>6</sup>

$$M = \sum_{\psi} P(\psi) M(\psi), \qquad (1)$$

where  $P(\psi)$  is the probability that a particular, fixed site on the lattice belongs to a finite <u>cluster</u>  $\psi$ -a group of  $\nu(\psi)$  occupied lattice sites with the property that all sites in  $\psi$  are connected to one other through a set of exchange bonds between nearest neighbors, and are not so connected to any occupied sites outside  $\psi$ -and  $M(\psi)$  is the magnetization of the cluster  $\psi$  divided by  $\nu(\psi)$ .

As we are concerned with Ising ferromagnets, the theorem of Lee and Yang<sup>8</sup> allows us to write  $M(\psi)$  in the form

$$M(\psi) = 1 + \frac{2z}{\nu(\psi)} \sum_{i} \frac{1}{\xi_{i} - z},$$
 (2)

where  $z = \exp(-2H/kT)$  and the  $\xi_i$ ,  $i = 1, 2, \dots$ ,  $\nu(\psi)$ , are complex numbers with  $|\xi_i| = 1$ . Consequently *M* itself is of the form 1 + zf(z) where

$$f(z) = \sum \eta_i [\xi_i - z]^{-1}$$
(3)

with  $\eta_i = 2P(\psi)/\nu(\psi)$  if  $\xi_i$  is one of the poles of  $M(\psi)$ . Note that  $\sum \eta_i = 2\sum P(\psi) = 2p$ , and thus (3) is absolutely convergent and defines an analytic function for |z| < 1. In addition, f(z) cannot be analytically continued through any point of the unit circle which is an accumulation point of the  $\xi_i$ . For suppose that  $z_1$ ,  $|z_1| = 1$ , is an accumulation point of the  $\xi_i$  and suppose there were a function  $\varphi(z)$  analytic inside a small circle C of radius R centered at  $z_1$  with the property that  $\varphi(z) = f(z)$  in the intersection of C and |z| < 1. Choose  $\xi_k$  such that  $|\xi_k - z_1| < R$  and consider the variation of f(z) on a radius drawn from 0 to  $\xi_k$ , 0 < r < 1:

$$f(r\xi_k) = \frac{\eta_k}{(1-r)\xi_k} + \sum_{i \neq k} \frac{\eta_i}{\xi_i - r\xi_k}.$$
(4)

If without loss of generality we assume that  $\xi_i \neq \xi_k$ for  $i \neq k$  (otherwise we would simply define a new  $\eta_k$  combining all contributions from the pole at  $\xi_k$ ), it is easy to place a bound on the second term in (4) so that it is in magnitude less than half the first term for r sufficiently close to 1; consequently  $f(r\xi_k)$ —and therefore  $\varphi(r\xi_k)$ —diverges as  $r \rightarrow 1$ , contrary to our assumption that  $\varphi$  is analytic.

To complete the argument for nonanalyticity of M at H = 0 (z = 1), consider a sequence of "regular" clusters which have the property that they consist of all sites in the lattice inside, but no sites outside, squares or cubes (for two- and three-dimensional lattices, respectively) of increasing size. Such clusters occur in the sum (1) with finite probability, and for  $T < T_c(1)$  the minimum of  $|\xi_i - 1|$  for a regular cluster goes to zero as the size goes to infinity,<sup>8</sup> since this is one way of obtaining the thermodynamic (infinitevolume) limit for the regular system. Consequently in this temperature range, z = 1 (H = 0) is an accumulation point of the  $\xi_I$  and *M* fails to be an analytic function at H = 0. For  $p \ge p_0$ , the above argument must be modified by adding to (1) a contribution from the "infinite cluster," but the conclusion remains unaltered.

Gallavotti<sup>9</sup> has obtained an upper bound for  $T_c^*$  for systems of the sort we are considering, while

our argument provides a lower bound,  $T_c(1)$ . It is a plausible expectation that for any p,  $T_c^*$ = $T_c(1)$ . There need be no "visible" change in the magnetization curves at  $T = T_c^*$ ; they could still be infinitely differentiable. On the other hand, it seems possible that for  $p > p_0$  one might find the susceptibility diverging at a temperature above that at which spontaneous magnetization first appears.<sup>10</sup>

There are some additional possible applications of our arguments. For example, at H = 0 the energy of a finite cluster has a representation analogous to (2) but with z defined as  $\exp(-2J/kT)$ , and the location of the  $\xi_i$  is not precisely known. If  $z_c$  (0 <  $z_c$  < 1) is a point of nonanalyticity for the energy of the regular system in the thermodynamic limit, it will also be a point of accumulation of the  $\xi_i$  in (3) with p < 1, and additional points of accumulation will appear between 0 and  $z_c$ . Unfortunately, since the  $\xi_i$  are not (so far as we know) restricted to lie on a particular curve or set of curves, we are unable to prove that f(z) lacks an analytic continuation through the points of accumulation,<sup>11</sup> The calculations of Mikulinskii<sup>12</sup> (again, not a rigorous proof) indicate that in the two-dimensional Ising model the energy of a random system fails to be analytic at  $T = T_c(1)$ , which concurs with our expectations.

The absence of definite theorems on the location of the appropriate  $\xi_i$  prevents us from making precise statements about random Ising antiferromagnets or Heisenberg ferromagnets (or antiferromagnets), though it is plausible that singularities in the regular system persist in the random system as nonanalyticities. Our results do not apply in a straightforward fashion to the random Ising ferromagnet considered by McCoy and Wu<sup>13</sup> because it does not have a simple decomposition into clusters. On the other hand, it is not unreasonable to expect that something analogous may be going on in this model, and a nonanalyticity in the heat capacity at the critical temperature of the regular system seems a distinct possibility, even though it has not thus far shown up in calculations.

\*Research supported in part by the U. S. Atomic Energy Commission.

<sup>†</sup>Permanent address: Physics Department, Carnegie-Mellon University, Pittsburgh, Pa. 15213.

<sup>1</sup>In the case of Ising ferromagnets this conjecture, though it has not yet been proved, is strongly supported by several rigorous results: see C. N. Yang and T. D. Lee, Phys. Rev. <u>87</u>, 404, 410 (1952); J. L. Lebowitz and O. Penrose, Commun. Math. Phys. <u>11</u>, 99 (1968); G. Gallavotti, S. Miracle-Sole, and D. W. Robinson, Phys. Letters <u>25A</u>, 493 (1967).

<sup>2</sup>For precise details of the random model, see R. B. Griffiths and J. L. Lebowitz, J. Math. Phys. <u>9</u>, 1284 (1968).

<sup>3</sup>For simplicity we restrict ourselves to a simple case, but our results may be easily generalized to (a) the situation where all sites are occupied, but exchange "bonds" between neighboring sites are removed at random, and (b) the case where the interaction is not limited to nearest neighbors, but is still zero beyond some finite range.

<sup>4</sup>I. Syozi, Progr. Theoret. Phys. (Kyoto) <u>34</u>, 189 (1965); I. Syozi and S. Miyazima, <u>ibid</u>. <u>36</u>, 1083 (1966).

<sup>5</sup>G. S. Rushbrooke and D. J. Morgan, Mol. Phys. <u>4</u>, 1, 291 (1961).

<sup>6</sup>A number of results will be domonstrated in detail and with suitable rigor in a longer paper now in preparation.

<sup>7</sup>M. E. Fisher, Phys. Rev. <u>162</u>, 480 (1967).

<sup>8</sup>Yang and Lee, Ref. 1.

<sup>9</sup>G. Gallavotti, to be published.

<sup>10</sup>There is indication of this behavior in a model studied by McCoy (private communication).

<sup>11</sup>One can construct examples with a suitable choice of  $\eta_i$  and  $\xi_i$  such that f can be continued analytically into regions containing points of accumulation of the  $\xi_i$ .

<sup>12</sup>M. A. Mikulinskii, Zh. Eksperim. i Teor. Fiz. <u>53</u>, 1071 (1967) [translation: Soviet Phys.-JETP <u>26</u>, 637 (1968)].

<sup>13</sup>B. McCoy and T. T. Wu, Phys. Rev. <u>176</u>, 631 (1968).