Droplet Model for Tricritical Points: Metamagnetic Transition*

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The phase transition of a two-sublattice spin-$\frac{1}{2}$ Ising antiferromagnet in an external magnetic field is studied on the basis of a modified form of Fisher's droplet model for critical phenomena. It is assumed that the interaction also contains an intrasublattice ferromagnetic coupling so that the model is appropriate for metamagnetic substances. By taking into account the contribution to the free energy, both of antiferromagnetic (AF) and of ferromagnetic (F) clusters, it is found that the phase boundary of the AF phase consists of a line of critical points at small fields when the AF clusters first become critical, but this line merges into a line of a first-order phase transition for magnetic fields above a threshold value H_t when the F clusters first become energetically favorable. The end point at (H_t, T_t) of the first-order phase transition is a tricritical point, in agreement with the results of the Landau and of the molecular-field theories. Two kinds of critical behavior at the tricritical point are found. In what is called a tricritical point of the first kind, only the AF clusters become critical at (H_t, T_t). In this case, the difference $M_t - M_-$ of the magnetizations of the two coexisting phases on the first-order transition goes to zero linearly at the tricritical point. The free energy does not have a homogeneous form near this point. At a tricritical point of the second kind, both AF and F clusters become critical. In this case it is found that $(M_+ - M_-) \sim |(T - T_t)/T_t|^{\zeta}$, where, in general, ζ is different and less than unity. Under certain conditions it is found that the free energy has a homogeneous form near (H_t, T_t). The critical behavior near (H_t, T_t) is discussed in detail for both kinds of tricritical points. Better agreement with experiments is found than in the case of the molecular-field theory. The tricritical point of dysprosium aluminum garnet seems to be of the second kind, but more detailed measurements are needed. The two-fluid critical mixing point in He^3-He^4 is briefly discussed.

I. INTRODUCTION

Detailed experimental information[1-3] is becoming available on the magnetic phase transition of metamagnets.[1,4] This name refers to strongly anisotropic antiferromagnets with the following behavior in an external magnetic field $\vec{H}$. If $\vec{H}$ is pointing along an appropriate direction (the easy axis for a two-sublattice antiferromagnet), the low-temperature antiferromagnetic phase ends with a continuous phase transition (the antiferromagnetic transition) for small fields $\vec{H}$ but with a first-order transition for fields larger than a threshold H_t. This first-order phase transition, which we call the metamagnetic transition, takes place between a phase with antiferromagnetic order and a paramagnetic phase of high magnetic moment induced by the external field. Therefore, the phase boundary of the antiferromagnetic phase in the H-T plane consists of a line of critical points which continues at (H_t, T_t) in a line of a first-order phase transition. The over-all qualitative behavior of a metamagnet is understood on the basis of the molecular-field theory for a strongly anisotropic magnetic Hamiltonian with an antiferromagnetic interaction between spins on different sublattices and a ferromagnetic interaction between spins on the same sublattice.[4,5] Similar results are obtained on the basis of Landau's theory.[6] However, we do not expect such "classical" theories to give a correct description of the critical phenomena near the continuous transition because we know that "classical" theories fail near a critical point.[7] On the other hand, the critical behavior in the neighborhood of (H_t, T_t), the end point of the first-order phase transition, is expected to be of particular interest. In fact, it has been recently pointed out by Griffiths[8] that this end point is an example of a tricritical point—a point which lies on the intersection of three lines of critical points. One of these lines corresponds to the antiferromagnetic transition line and the two others are lines at which two coexistence surfaces of another first-order transition terminate. These two surfaces extend in the $H_s \neq 0$ region and their intersection with the $H_s = 0$ plane coincides with the metamagnetic line. H_s is the staggered magnetic field, that is, a magnetic field which points in opposite directions on the two magnetic sublattices.

Experiments on metamagnets, as well as on He^3-He^4 mixtures near their two-fluid critical mixing point, which is another example of a tricritical point, show some discrepancies with the predictions of the "classical" theory.[8] Moreover, some metamagnets and He^3-He^4 mixtures seem to differ, to a certain extent, in the critical behavior near their tricritical point. All this indicates the necessity of going beyond the "classical" theory. Griffiths has proposed[8] a homogeneous form for the

free energy in the neighborhood of a tricritical point which resolves the discrepancies in the case of He^3-He^4 mixtures. However, we should like to have a description based on more physical terms which might also help to understand if the characterization of different tricritical points is unique or not. To this end we have studied the phase transition of a metamagnet on the basis of a generalization of Fisher's droplet or cluster model for critical phenomena.[9] This model is particularly attractive because in the case of an ordinary critical point it goes beyond the "classical" theory by giving free energy which agrees with the results of the scaling description of critical phenomena[7] and, on the other hand, it is formulated in a form which is physically rather transparent. We construct the theory for a spin-$\frac{1}{2}$ Ising model, but the theory also describes the behavior of a strongly anisotropic Heisenberg model such that no "spin-flop" phenomenon[10] occurs. Therefore, our model should give a description of the metamagnetic compounds.

The basic idea of the cluster model can be formulated in the following form in the case of an antiferromagnet: If we compare the free energy of the system at a temperature T and in a staggered field H_s with the free energy of the system at the same temperature, but in the completely ordered state (this can be obtained by $H_s \to \infty$), the main contribution to the difference between these two free energies comes from antiferromagnetic clusters of compact shape—regions of overturned spins so that the sublattice magnetization has locally the wrong direction. In this case, one is able to estimate the contribution of a cluster to the free energy and its probability of occurrence, if one neglects the interaction between clusters. To extend the model, in case an external magnetic field is present, we must also take into account the contribution from ferromagnetic clusters. We find that if the interaction between spins on the same sublattice is ferromagnetic, these ferromagnetic (F) clusters have a positive surface free energy at low temperatures, like the antiferromagnetic (AF) clusters. Therefore, also F clusters have a compact shape and we are able to evaluate their relative contribution to the free energy. By suitable population factors we also take into account the "exclusion" effects between clusters of the same type and between AF and F clusters. In fact, we have recently argued,[11] in the case of an ordinary critical point, that to have a more realistic description of the system above the transition temperature, the droplet formula should also contain an explicit population factor which takes into account the "exclusion" effect between clusters. Explicit calculations[12] with this modified droplet formula show that it gives a rather good description of the critical behavior of the Ising ferromagnet in two and three dimensions by also giving the critical amplitudes of significant thermodynamic quantities in fair agreement with the exact results or with the results of series analysis.[7]

Our formula for an Ising antiferromagnet in an external magnetic field contains two phase boundaries in the H-T plane ($H_s = 0$): One is the locus on which the surface free energy of AF clusters is zero, and this corresponds to the AF transition. The other boundary corresponds to the intersection with the $H_s = 0$ plane of a surface extending in the $H_s \neq 0$ region. This surface gives the limit of stability of the AF phase with respect to F clusters and this represents a surface of coexisting phases of a first-order phase transition. We identify this second boundary in the H-T plane with the metamagnetic transition, and the tricritical point corresponds to the intersection of the two boundaries. The fluctuations in the neighborhood of the tricritical point can be quite different, depending on the magnitude of T_t with respect to T^*, the temperature at which the microscopic surface free energy of F clusters is zero. If $T^* > T_t$, only the AF clusters become critical at the tricritical point. In this case we find that the difference ΔM of the magnetizations of the two coexisting phases at the metamagnetic transition goes to zero linearly with $T_t - T$. If $T^* = T_t$, both F and AF clusters become critical at the tricritical point, and in this case ΔM goes to zero with a power behavior $|T - T_t|^{\zeta}$ with an exponent ζ, which is most likely less than unity. It seems probable that the parameters of the theory are never such that $T^* < T_t$ because, in this case, the interaction between the two different kinds of clusters has the tendency to depress the AF boundary until $T^* = T_t$. Not only ΔM has a different behavior near (H_t, T_t) depending upon whether $T^* > T_t$ or $T^* = T_t$, but the whole critical behavior is different in the two cases. We speak of a tricritical point (and, by extension, a metamagnet) as being of first or second kind corresponding to $T^* > T_t$ or $T^* = T_t$, respectively. In the case of a tricritical point of the first kind, the free energy does not have a homogeneous form, contrary to Griffiths's proposal.[8] If suitable restrictions are satisfied by the parameters of the model the free energy has a homogeneous form near a tricritical point of the second kind. This homogeneous form coincides with the one proposed by Griffiths if the basic exponents of the model are such that $\zeta = 1$ results. We also construct a droplet formula for the paramagnetic high-field phase, starting, in this case, from the ferromagnetically ordered state present when $H \to \infty$. In this way we are also able to describe the paramagnetic phase at the metamagnetic transition. The droplet formula for the free energy has been constructed in

the case of a magnetic system. However, we expect that the formula also applies in the case of other tricritical points whenever the ordered phase is destroyed by two different and competing kinds of fluctuations, both of which are characterized by a positive microscopic surface free energy at small temperatures.

At least one metamagnet,[3] dysprosium aluminum garnet (DAG), has a power behavior for ΔM with $\zeta < 1$, thus suggesting that this compound is an example of a metamagnet of second kind. The experimental data, however, are not yet detailed enough to exclude the possibility of a very steep linear behavior for ΔM. The difference of composition, a quantity analogous to ΔM, has a linear behavior near the two-fluid critical mixing point[13] of He^3-He^4. Therefore, He^3-He^4 mixtures seem to offer an example of a tricritical point of the first or second kind but in the exceptional case in which $\zeta = 1$.

The contents of the paper are the following. In Sec. II we construct the free energy for the model and in Sec. III the surface energy of clusters is studied for two different AF structures: the case of two compenetrating cubic lattices[3] and the case of antiferromagnetic stacking of ferromagnetically ordered planes.[1,2] The metamagnet of first and of second kind is studied in Secs. IV and V, respectively. In Sec. VI the renormalized critical behavior[14] near the tricritical point is discussed, and, finally, Sec. VII contains a comparison of the theory with experiments and the conclusions.

II. DROPLET FORMULA

We consider a spin-$\frac{1}{2}$ Ising system with Hamiltonian

$$\mathcal{H} = 2J_{AF} \sum_{\langle i\mu \rangle} s_i^z s_\mu^z - 2J_F \left(\sum_{\langle ij \rangle} s_i^z s_j^z + \sum_{\langle \mu\nu \rangle} s_\mu^z s_\nu^z \right) + (H - H_s) \sum_i s_i^z + (H + H_s) \sum_\mu s_\mu^z , \quad (1)$$

where the Latin and the Greek indices refer to sites on the a ("up") and b ("down") sublattices, respectively. $\langle i\mu \rangle$ indicates a sum over pairs of nearest neighbors on different sublattices and $\langle ij \rangle$, $\langle \mu\nu \rangle$, a sum over pairs of nearest neighbors on the same sublattice. H and H_s represent, respectively, a local magnetic field and a staggered magnetic field in the z direction in units of $g\mu_B$. The positive quantities J_{AF} and J_F must be consistent with the assumed AF order of the system in its ground state when $H = H_s = 0$.

On basis of the mean-field theory,[5,8] we expect that the phase diagram of such a system has in the (T, H, H_s) space the shape given in Fig. 1, where only the $H_s > 0$, $H > 0$ region is shown. There is an obvious symmetry for change of sign of H and of H_s if the two sublattices are equivalent. The phase diagram has two regions of coexisting phases. A first region lies on the $H_s = 0$ plane and the coexisting phases are characterized by (M_s, M) and by $(-M_s, M)$, where M_s and M are the staggered and the total magnetization as follows:

$$M_s = M_a - M_b , \quad M = M_a + M_b , \quad (2)$$

where M_a and M_b are the magnetizations of the two sublattices. The boundary of this AF region consists of two parts. Curve 1 corresponds to the AF phase transition where $M_s \to 0$, and it is characterized by a field-dependent transition temperature $T_N(H)$. Curve 2 corresponds to the first-order metamagnetic transition where three phases coexist, two AF, (M_s, M_-) and (M_s, M_-), and one F, $(0, M_+)$. This part of the phase boundary, characterized by a field-dependent temperature $T_m(H)$, represents the intersection with the $H_s = 0$ plane of another surface of coexisting phases which differ, this time, both for the values of M_s and of M. The differences in M and in M_s vanish on curve 3 which corresponds, therefore, to a line of continuous phase transitions. The tricritical point (H_t, T_t) lies at the intersection of three lines of critical points: 1, 3, and the symmetric of the last one in the negative H_s region.

The cluster model gives the free energy as a high-field (H_s for an antiferromagnet) expansion, that is, a power expansion in the variable $\exp(-H_s/K_B T)$. This expansion can be obtained if in the partition function

$$Z = \sum_{(s_i, s_\mu)} \exp[-\mathcal{H}(s_i, s_j)/K_B T]$$

one singles out the term corresponding to the perfect AF order (the only important term when $H_s \to \infty$) and classifies the other terms on the basis of the number of spins deviating from that order. These overturned spins can be collected in clusters and we expect that near the phase transition only rather large clusters are important.[9] We review briefly the usual droplet model by considering the $H = 0$ case first. The spin configurations which enter, with some weight, the partition function correspond to the presence of regions in which the spins are antiferromagnetically ordered but with an interchange of the roles of the a and b sublattices. If one neglects the interaction between these AF clusters and assumes that the clusters are compact, then the free energy for spin takes the well-known cluster form:

$$F^0(T, H_s) \approx -\tfrac{1}{2} H_s - U_0 - K_B T \sum_{l=1}^{\infty} g_1(\bar{s}_1) \exp[-\mathcal{S}_1(l)/K_B T] , \quad (3)$$

where U_0 is the interaction energy in the fully or-

dered state and $\mathcal{E}_1(l)$ is the difference between the energy of an AF cluster of l spins and the energy of the same region in the fully ordered state. For large clusters, $\mathcal{E}_1$ has the form

$$\mathcal{E}_1(l) = H_s l + \bar{s}_1 w_1 , \tag{4}$$

where the last term is a surface energy due to the broken bonds at the surface of the cluster which is assumed to have an average area $\bar{s}_1 = \bar{a}_1 l^{\sigma}$. The combinatorial factor $g_1(\bar{s}_1)$ is proportional to the number of AF clusters of l spins with a fixed center and surface $\bar{s}_1$, and it has the form[9] $g_1(\bar{s}_1) \simeq g_1 \lambda_1^{\bar{s}_1} \times \bar{s}_1^{\tau/\sigma}$, where g_1, λ_1, and τ are constants. Then (3) can be written in the form

$$F^0(T, H_s) = -\tfrac{1}{2} H_s - U_0 - q_0 K_B T \sum_{l=1}^{\infty} l^{-\epsilon} Y_1^l X^{l^{\sigma}} f(l) , \tag{5}$$

where

$$Y_1 = \exp(-H_s/K_B T) , \tag{6}$$

$$X = \lambda_1^{\bar{a}_1} \exp(-\bar{a}_1 w_1/K_B T) = \exp[-\bar{a}_1 (w_1 - \bar{\omega}_1 T)/K_B T] , \tag{7}$$

and we have written $\lambda_1 = \exp(\bar{\omega}_1/K_B)$. In (5) we have also introduced a population factor $f(l)$. If $f(l) = 1$, (5) is Fisher's droplet formula. We have argued[11] for the presence of such a population factor on the basis that only compact clusters contribute to the singular part of the free energy and, moreover, that the presence of a cluster on some sites excludes other clusters from the same sites. The form

$$f(l) = (B X^{b l^{\sigma}} + 1)^{-1} , \quad b > 1 \tag{8}$$

was suggested[11] and found satisfactory for the Ising model.[12] On the basis of expression (5) for the free energy, the following picture of the phase transition emerges. As long as $H_s > 0$ ($Y_1 < 1$) the fluctuations, as described by (5), are finite. Also at $H_s = 0$ ($Y_1 = 1$) and at low temperatures, such that $X < 1$, the fluctuations remain finite but they increase with T up to $T = T_N = w_1/\bar{\omega}_1$. At this temperature the fluctuations are infinite, as shown by the fact that the derivatives of F beyond a certain order diverge, and this point is identified with the Néel temperature. For $T > T_N$ the "microscopic" surface free energy $(w_1 - \bar{\omega}_1 T)$ is negative and the AF order is no longer spontaneously stable. One does not automatically find $M_s = 0$ when $H_s = 0$ because (5) does not possess the symmetry that the total free energy has under $H_s \to -H_s$. B and b in (8) must be chosen[11] in such a way that $M_s = 0$ results when $H_s = 0$ and $T > T_N$. For $H_s < 0$ the series (5) diverges, and this corresponds to the fact that a state with a negative value for M_s is stable. From (5) it is easy to calculate[9,11] the critical exponents and, in particular, they turn out to be equal above and below T_N.

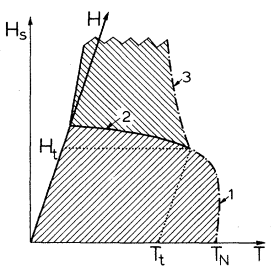

FIG. 1. Schematic phase diagram for a metamagnet in T, H, H_s space. The hatched areas represent surfaces of coexisting phases (first-order transition). Curves 1 and 3 are lines of critical points.

In the presence of an external magnetic field, the free energy still contains the AF contribution (3), but now the surface energy w_1 depends on H and, at least for small fields, we have

$$w_1(H) = w_1 - AH^2 , \tag{9}$$

where A is a constant. However, in this case we expect that configurations containing regions of ferromagnetically ordered spins also give an important contribution to the free energy. In Sec. III we show that these F clusters also have a positive surface energy, so that we may assume that they have a compact shape. Neglecting again the interaction between clusters, the free energy contains another contribution which coincides with (3) in form, but with functions $g_2(\bar{s}_2)$ and $\mathcal{E}_2(l)$. The combinatorial factor g_2 has the same form as g_1, but it could have different constants g_2, λ_2, and τ_2 because the average area for F clusters might be characterized by different constants $\bar{a}_2$ and σ_2 so that $\bar{s}_2 = \bar{a}_2 l^{\sigma_2}$. The cluster energy $\mathcal{E}_2(l)$ contains again a bulk and a surface term and it has the form

$$\mathcal{E}_2(l) = [E_2(H, H_s, T) - \tfrac{1}{2}(H - H_s)]\, l + \bar{a}_2 w_2 l^{\sigma_2} . \tag{10}$$

The bulk term contains the factor $\frac{1}{2}(H - H_s)$ because a F cluster of l spins is constructed by overturning only the spins of the b sublattice. $E_2(H, H_s, T)$ is the difference between the interaction energy of the spins in the F and AF configurations. Actually, we expect that this energy depends on H, H_s, and T because it should be the difference of the thermal averages in the two different constrained configurations. At finite temperatures we expect that the energy E_2 decreases for an increase of T or H, whereas we expect the opposite dependence on H_s. When necessary, we will use the following form:

$$E_2(H, H_s, T) = E_2 - B' T^2 (1 + CH^2 - FH_s^2) ,$$

where E_2, B', C, and F are constants. However, we notice that no results depend on this particular form of E_2 as long as it is an analytic function of its variables.

On the basis of the previous discussion, it follows that the significant part of the free energy per spin can be written in the form

$$F(T, H, H_s) \approx -\tfrac{1}{2} H_s - U_0 + F_1(T, H, H_s) + F_2(T, H, H_s) , \quad (11)$$

where

$$F_1(T, H, H_s) = -q_1 K_B T \times \sum_{l_1=1}^{\infty} l_1^{\tau_1} Y_1^{l_1} X_1^{\lambda_1(l_1)} f_{11}(l_1) f_{12}(l_1) , \quad (12)$$

$$F_2(T, H, H_s) = -q_2 K_B T \times \sum_{l_2=1}^{\infty} l_2^{\tau_2} Y_2^{l_2} X_2^{\lambda_2(l_2)} f_{21}(l_2) f_{22}(l_2) , \quad (13)$$

$$\lambda_1(l_1) = l_1^{q_1}, \quad \lambda_2(l_2) = l_2^{q_2} , \quad (14)$$

$$X_1 = \exp\{-\bar{a}_1[w_1(H) - \bar{\omega}_1 T]/K_B T\} , \quad (15)$$

$$Y_2 = \exp\{-[E_2(H, H_s, T) - \tfrac{1}{2}(H - H_s)]/K_B T\} , \qquad X_2 = \exp\{-\bar{a}_2(w_2 - \bar{\omega}_2 T)/K_B T\} , \quad (16)$$

and Y_1 is given in (6). The index 1 refers to AF clusters and the index 2 refers to F clusters. In (12) and (13) we have introduced the population factors $f_{ij}(l)$. For $f_{11}(l)$ and $f_{22}(l)$, which give the effect of AF and of F clusters on themselves, we use the form[11]

$$f_{11}(l) = (B_1 X_1^{b_1 \lambda_1(l)} + 1)^{-1} , \quad f_{22}(l) = (B_2 X_2^{b_2 \lambda_2(l)} + 1)^{-1} , \quad (17)$$

where B_1, B_2, b_1, and b_2 are constants. Analogous factors take into account the effect of F clusters on AF clusters and vice versa:

$$f_{12}(l) = (B_{12} X_2^{b_{12} \lambda_2(l)} Y_2^{l} + 1)^{-1} , \qquad f_{21}(l) = (B_{21} X_1^{b_{21} \lambda_1(l)} Y_1^{l} + 1)^{-1} . \quad (18)$$

These terms significantly differ from (17) by the presence of the factors Y_2^l and Y_1^l, respectively. The reason for this difference is the following. Let us consider the population effect on a AF cluster. For example, a cluster of l spins might have a very irregular surface and in this case it will not contribute to F_1. The probability of occurrence of such a cluster depends on $X_1^{\lambda_1(l)}$, being very large when $X_1 > 1$ and becoming larger as the surface of the cluster increases. This corresponds to the behavior of f_{11} given in (17) which should give the probability that a cluster is compact. On the other hand, Y_1 should not enter f_{11} because we started with a cluster of l spins and, whatever the shape of its boundary, this gives a factor Y_1^l which appears explicitly in (12). The situation is different with respect to the factor f_{12} because, in this case, one is interested in the probability that an AF cluster is irregular owing to inclusion of F clusters. The Y_1^l factor in (12) does not give any constraint on F clusters because these have an equal number of "up" and "down" spins. On the other hand, the probability of occurrence of F clusters is large or small depending on whether $Y_2 > 1$ or $Y_2 < 1$. This effect is taken into account by the form for f_{12} given in (18). A similar argument applies to the factor f_{21}. We notice that the precise form of f_{ij} given in (17) and (18) does not matter as long as f_{ij} depend on the same arguments and have the same limits when $l \to \infty$. Moreover the constants appearing in (12), (13), (18), and (19) might be analytic functions of T, H, and H_s.

The AF order is stable as long as $Y_1 < 1$ and $Y_2 < 1$ and becomes unstable when one of these two variables becomes greater than the unity. If, for fixed H and T, the surface defined by $Y_1 = 1$ is first met, the instability is due to the formation of a phase with dominant AF order with the roles of a and b interchanged. On the other hand, if the surface $Y_2 = 1$ is first met, the instability is due to the formation of a phase with dominant F order. Therefore, the region of the plane $H_s = 0$ ($Y_1 = 1$) with $Y_2(H, 0, T) < 1$ gives a possible surface of coexisting AF phases of a first-order phase transition. This region actually represents a first-order phase transition only if $X_1 < 1$. The region of coexisting phases terminates on a line of critical points where $X_1 = 1$, corresponding to a field-dependent transition temperature $T_N(H)$, where

$$T_N(H) = w_1(H)/\bar{\omega}_1 \underset{H \to 0}{\sim} T_N - A' H^2 . \quad (19)$$

The other possible surface of coexisting phases, one with dominant AF order and the other with dominant F order, is determined by $Y_2 = 1$ or

$$E_2(H, H_s, T) = \tfrac{1}{2}(H - H_s) , \quad (20)$$

which can be solved for T to give a function $T_2(H, H_s)$. Only the region of this surface with $X_2 < 1$, i.e., $T_2(H, H_s) < T^*$, where

$$T^* = w_2/\bar{\omega}_2 , \quad (21)$$

corresponds to a first-order transition.

Let us call $T = T_m(H) \equiv T_2(H, 0)$ the intersection curve between the surface given by Eq. (20) and the $H_s = 0$ plane and let us call (H_t, T_t) the point at the intersection of the two curves $T = T_N(H)$ and $T = T_m(H)$, i.e.,

$$T_t = T_N(H_t) = T_m(H_t) . \quad (22)$$

The behavior near (H_t, T_t) is rather different depending on the magnitude of T^* with respect to T_t. If $T^* > T_t$ [see Fig. 2(a)] at the point (T_t, H_t), the AF clusters are critical, that is, their surface free energy is zero, so that there is a large probability of finding large clusters, whereas the F clusters are normal. Also, if F clusters are normal, this point (H_t, T_t) is the end point of the metamagnetic transition and it can be identified with the tricriti-

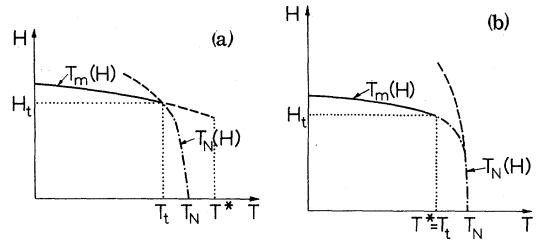

FIG. 2. Schematic H, T phase diagram. Solid line represents metamagnetic transition; dot-dashed line represents antiferromagnetic transition. (a) $T_t < T^*$. Dashed lines give the continuation of $T_N(H)$ and $T_m(H)$ but do not represent lines of phase transition. (b) $T_t = T^*$. Dashed line represents the unrenormalized $T_N(H)$.

cal point in Fig. 1. In fact, as one moves along the $T_m(H)$ line in the direction of higher temperatures, first F clusters become larger because the surface free energy of these clusters becomes smaller. However, beyond T_t the opposite behavior occurs because $X_1 > 1$ there and the population factor f_{21} in (13) limits the size of the F clusters. At T^* all the derivatives of F_2 are finite because the factor f_{21} assures convergence of the series (13) for F_2. This corresponds to the fact that even if near T^* the surface free energy of the F clusters is small, there is a vanishing probability of finding large and compact F clusters because there the AF clusters are overcritical, so that there is a very large number of them.

If $T^* = T_t$, at the point (H_t, T_t) both the AF and the F clusters become critical. The case $T^* < T_t$ [see Fig. 2(b)] does not seem to take place. In fact, suppose this was the case and let us consider the system near T_t. Very large AF clusters are present because one is near $T_N(H)$. In Sec. III we show that the surface energy of an AF cluster is decreased by decorating it with small F clusters. On the other hand, there is a large probability that this happens because $Y_2 = 1$ and $X_2 > 1$, so that there is a very large number of these small F clusters. The over-all effect of this interaction between clusters is that of decreasing $T_N(H)$, and it seems likely that this renormalization of $T_N(H)$ is such that $T^* = T_t$. Next we study the $T^* = T_t$ case and we do not consider the other possibility, namely, $T^* < T_t$. One might also wonder if, in the case $T^* > T_t$, the interaction between clusters should not cause $T^* = T_t$. This does not seem to be the case because decoration of a large F cluster with small AF clusters does not change the surface energy, at least for the models considered in Sec. III.

As we said before, for $H_s \neq 0$, the surface $T = T_2(H, H_s)$ represents a surface of coexisting phases which terminates at $T = T^*$ on a line of critical points. Therefore, if $T^* = T_t$, the point (T_t, H_t) is a tricritical point because it lies at the intersection of three lines of critical points. This is not the case if $T^* > T_t$ because on the basis of (11) the first-order phase transition terminates at T_t if $H_s = 0$ but it terminates at T^* if $H_s \neq 0$. This follows from the fact that

$$\lim_{l \to \infty} f_{21}(l) = 0 \quad \text{if } H_s = 0$$

and

$$\lim_{l \to \infty} f_{21}(l) = 1 \quad \text{if } H_s \neq 0 .$$

This discontinuous behavior is not realistic and it can be traced to a defect of the form (18) for the population factor. Consider, for example, the effect of F clusters upon themselves. A cluster of l spins is excluded by the other clusters of any size, whereas Eq. (17) for $f_{22}(l)$ depends only on $X_2^{\lambda_2 (l)}$. The reason for this choice is that the population factor plays an essential role only when the clusters are overcritical ($X_2 > 1$), and in this case the largest probability is for clusters of size l. So we have approximated the exclusion effect by its largest term. A similar situation applies to $f_{21}(l)$, with respect to the effect of AF clusters on F clusters, when $X_1 \geq 1$ and $Y_1 \geq 1$. On the contrary, if $X_1 > 1$ and $Y_1 < 1$, the probability for AF clusters first increases with l, owing to the $X_1^{\lambda_1 (l)}$ factor, but it finally decreases when the Y_1^l factor takes over. If H_s is very small, the total number of AF clusters is very large and the exclusion effect on F clusters is important, whereas the approximation (18) for $f_{21}(l)$ severely underestimates this effect, particularly if l is large. A more realistic form for $f_{21}(l)$ would be a function which in the $l \to \infty$ limit is equal to 1 or to 0, depending on whether $P > q_c$ or $P < q_c$. P is the fraction of available sites for F clusters and q_c is a constant expected to be related to the critical percolation probability for the given lattice.[15] We do not discuss further a more realistic form for f_{21} because we are interested only in the case $H_s = 0$, the experimentally accessible plane. We simply assume that with a more realistic form for f_{21} the first-order phase transition terminates on a line on the surface $T = T_2(H, H_s)$ which is continuous, ends at T_t on the $H_s = 0$ plane, and approaches T^* for large values of H_s. Thus, (T_t, H_t) is a tricritical point in this case also.

Summarizing the results of this section, we find that, for a given field H, the AF phase terminates with a continuous transition or with a first-order phase transition, depending on whether first the AF clusters become critical or first the F clusters become energetically favorable. For the tricritical point we find two possibilities. Either the metamagnetic transition terminates because the F clusters become critical as well as the AF ones, or the transition is driven by the fact that the AF

clusters become critical, whereas the F clusters remain normal. We expect a rather different critical behavior in the two cases, and this expectation is borne out by the calculations of Secs. IV and V.

If one is interested in the free energy of the high-field paramagnetic phase, in the expression for the partition function one should order the sum over spin configurations with respect to the fully ordered F configuration, the only state which contributes to F in the limit $H \to \infty$ and H_s finite. The main contribution to F comes, at least at small temperatures, from AF clusters. These clusters now play exactly the role the F clusters have with respect to the antiferromagnetic state. A difference is that the energy of an AF cluster has an expression similar to (10) but with the bulk term changed in sign, i.e.,

$$\tilde{\mathcal{E}}_2(l) = [\tfrac{1}{2}(H - H_s) - E_2(H, H_s, T)]\, l + \bar{a}_2 w_2 l^{\sigma_2} . \quad (23)$$

These clusters give a contribution $\tilde{F}_2$ to the free energy $\tilde{F}$, now written with a tilde to distinguish it from that of the previous case. We have

$$\tilde{F}(T, H, 0) \approx -\tfrac{1}{2}H - \tilde{U}_0 + \tilde{F}_1(T, H, 0) + \tilde{F}_2(T, H, 0) , \quad (24)$$

where $\tilde{U}_0$ is the interaction energy in the fully F ordered state (we consider only the $H_s = 0$ case). Taking into account Eq. (13), F_2 can be written in the form

$$\tilde{F}_2(T, H, 0) = -\tilde{q}_2 K_B T \times \sum_{l_2=1}^{\infty} l_2^{-\tau_2} Y_2^{-l_2} X_2^{\lambda_2 (l_2)} f_{22}(l_2) f_{21}(l_2) , \quad (25)$$

where Y_2 and X_2 are given by (16) and the population factors are given in (17) and (18). At the possible phase boundary $T = T_m(H)$, an AF cluster is, on the average, a single domain as long as $X_1 < 1$, that is, for $T_m(H) < T_t$. However, as we approach T_t, $X_1 \to 1$ and an AF cluster has the tendency to consist of many domains. In (24) we have included the term $\tilde{F}_1$, which takes into account these fluctuations inside the AF clusters. Since near T_t the total volume occupied by the AF clusters is a finite and rather large fraction of the total volume, we approximate $\tilde{F}_1$ by an expression similar to (12):

$$\tilde{F}_1(T, H, 0) = -\tilde{q}_1 K_B T \sum_{l_1=1}^{\infty} l_1^{-\tau_1} X_1^{\lambda_1 (l_1)} f_{11}(l_1)\, \tilde{f}_{12}(l_1), \quad (26)$$

where $\tilde{q}_1$ is a constant, $f_{11}(l)$ is given in (17), and $\tilde{f}_{12}(l)$ has the same form as f_{12} but with different constants $\tilde{B}_{12}$ and $\tilde{b}_{12}$:

$$\tilde{f}_{12}(l_1) = (\tilde{B}_{12} X_2^{\tilde{b}_{12}\lambda_1 (l_1)} Y_2^{l_1} + 1)^{-1} \quad (27)$$

because of the somewhat different role this term has in this case. On the basis of (24) we conclude that $T_m(H)$ is the temperature of the metamagnetic transition for $T_m(H) < T_t$, which is in agreement with our previous discussion. The different constants which appear in the expressions for F and $\tilde{F}$ must be such that the two free energies (11) and (24) coincide all along $T_m(H)$. Moreover, the entropies and the magnetizations calculated on the basis of (11) and (24) must coincide along $T_m(H)$ for $H < H_t$ because on this part of the curve there is no phase transition. This condition corresponds to the one imposed on (5) that $M_s = 0$ when $H_s = 0$ and $T > T_N$.

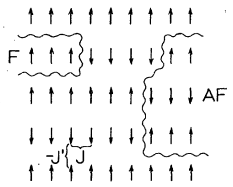

FIG. 3. Antiferromagnetic spin configuration with a F and AF cluster.

III. CLUSTERING PROPERTIES

The droplet formula for a metamagnet, given in Sec. II, is based on the existence of a clustering tendency for the spins which are overturned under the influence of the external field H to form a F cluster. To study this property one must specify the Hamiltonian of the system. In a family of compounds,[1,2] like $FeCl_2$, a sublattice consists of the spins on alternating planes and the basic part of the interaction consists of a F coupling $-J$ between nearest-neighboring (nn) spins on the same plane and of an antiferromagnetic coupling J' between nn spins on nn planes. For simplicity, we assume that the spins are located at the vertices of a simple tetragonal lattice. If the F planes are parallel to the xy plane, the spin distribution on a xz plane is shown in Fig. 3. Other compounds, like DAG, have a more complicated magnetic structure.[3] However, the system can be treated as a simple AF with two intercompenetrating sublattices if the magnetic field H is in particular directions. As a typical structure for this case, we consider a simple cubic lattice with nn AF coupling J_1, and second-neighbor ferromagnetic coupling $-J_2$. In Fig. 4 the spin distribution on one of the coordinate planes is shown.

We begin by considering the case of the layer structure (Fig. 3). In this figure we have also shown some examples of clusters. An AF cluster corresponds to a region of overturned spins equally distributed, on the average, on the two sublattices. There is clearly a clustering tendency because there is a positive surface energy corresponding to the "wrong" couplings of the spins at the boundary. Part of the boundary affects the J coupling and the remaining part the J' coupling. It is possible to calculate the shape of a cluster which

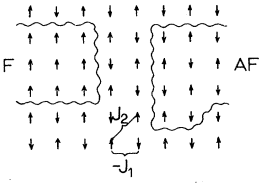

FIG. 4. As for Fig. 3 for a different antiferromagnetic configuration.

has the lowest energy. If a cluster has an extension of n_x, n_y, and n_z spins in the three coordinate directions, by minimizing the surface energy with the constraint that $n_x n_y n_z = \text{const}$, we find

$$n_x/n_y = 1\,,\quad n_z/n_x = J'/J\,,\quad w_1 = 6\,JJ'/(J+2J')\,, \tag{28}$$

where w_1 is the surface energy per spin. A F cluster corresponds to a region in which only the spins of the b sublattice are overturned. Since there is no direct coupling between spins of the same sublattice but on different planes, there is no clustering tendency with respect to the z axis. However, clustering is present inside a plane because of the broken J bonds, and we conclude that F clusters are two dimensional. Simple considerations give the bulk energy E_2 at zero temperature and the surface energy w_2, respectively, as

$$E_2 = 4J'\,,\quad w_2 = 2J\,. \tag{29}$$

Moreover, we expect that the exponent σ_2 for the average surface of a F cluster is close to $\frac{1}{2}$, the two-dimensional value, whereas σ_1 is expected to be closer to $\frac{2}{3}$, the three-dimensional value. However, any ferromagnetic interaction between spins on second-neighboring planes changes this two-dimensional character into a three-dimensional character. In case F clusters are truly two dimensional, the factor $\frac{1}{2}$ in front of $(H - H_s)$ in (10) must be deleted.

We consider now the interaction between AF and F clusters. We are interested in the effect of decorating the surface of a large cluster with small clusters of the other species. Between a large AF cluster and a small F cluster there is an attractive interaction because, if the F cluster is in contact with the boundary of the AF cluster, a smaller number of bonds are broken. If there is a finite probability of finding F clusters on the surface of an AF cluster, the resulting effect can be described as a decrease of the surface energy of AF clusters. On the contrary, since the F clusters are two dimensional, so that their boundary reduces to a line, the probability of contacts along this boundary with the AF clusters is very much reduced, and it seems likely that we may neglect the resulting effect of the interaction on w_2. Contacts of other kinds may renormalize the bulk term $E_2(H, H_s, T)$.

For the cubic antiferromagnet too, some examples of clusters are shown in Fig. 4. The surface energy of AF clusters has a positive contribution both from broken J_1 and J_2 bonds. The surface energy of F clusters does not depend on J_1 because the number of broken J_1 bonds does not depend on the configuration of the overturned spins of the b sublattice. On the contrary, the number of broken J_2 bonds depends on this configuration and one easily finds that the surface energy per spin is

$$w_2 = 8J_2\,. \tag{30}$$

We conclude that there is a clustering tendency if the second-neighbor interaction is ferromagnetic. Moreover, the geometry of F clusters is the same as that of AF clusters, so that we may expect not too dissimilar values for the exponents σ_1 and σ_2 and for τ_1 and τ_2. Should the second-neighbor interaction J_2 also be antiferromagnetic there would be an anticlustering tendency and our model would not be valid. We speculate that in this case the AF transition continues to $T = 0\,^\circ\text{K}$, where the transition becomes first order.

Also, in the case of the magnetic structure of Fig. 4 the interaction between F and AF clusters is asymmetric when one cluster is large and the other is small. In fact, it makes no difference for a small AF cluster to be at the surface of a F cluster because the number of broken J_1 and J_2 bonds is the same at the interface of these two clusters or at the interface of the F cluster with the original AF configuration. On the contrary, there is a difference in the case of a small F cluster on the surface of a large AF cluster. In fact, a F cluster is obtained by overturning some spins on the b sublattice. However, for those spins which are at the interface with the AF cluster the correct local order of the spins is restored. We conclude that it is convenient to decorate the surface of an AF cluster and this effect can be described again in terms of a reduction of its surface energy. The previous argument holds true only if the F clusters are rather small and there is a very large number of them. If this is not the case, the loss in the entropic factor becomes relatively more important than the energy gain.

IV. METAMAGNET OF FIRST KIND

In this section we study the critical behavior of a metamagnet of the first kind, i.e., a system for which $T^* > T_t$, where T^* and T_t are defined by (21) and (22), respectively. If $H < H_t$, the F_2 term of the free energy (11) is a regular function of its arguments and only the F_1 term has a "singular" part at the antiferromagnetic line $T_N(H)$. It is convenient to split Eq. (12) for F_1 in two parts:

$$F_1(T, H, H_s) = F_{1s}(T, H, H_s) + G(T, H, H_s)\,, \tag{31}$$

so that only F_{1s}, given by

$$F_{1s}(T, H, H_s) = -q_1 K_B T \sum_{l_1=1}^{\infty} l_1^{-\tau_1} Y_1^{l_1} X_1^{\lambda_1(l_1)} f_{11}(l_1) , \tag{32}$$

contains the "singular" part of the free energy. In fact, this term has the same structure as the usual droplet formula (5), so that some of the derivatives of this term are divergent at $Y_1 = 1$ ($H_s = 0$) and $X_1 = 1$ [$T = T_N(H)$]. The remaining term

$$G(T, H, H_s) = -q_1 K_B T \times \sum_{l_1=1}^{\infty} l_1^{-\tau_1} Y_1^{l_1} X_1^{\lambda_1(l_1)} f_{11}(l_1) [f_{12}(l_1) - 1] \tag{33}$$

is analytic at $Y_1 = 1$, $X_1 = 1$; this is guaranteed by the $(f_{12} - 1)$ factor. Taking into account Eqs. (6), (15), and (19), we see that (31) can be written as

$$F_{1s}(T, H, H_s) = \frac{T}{T^*(T, H)} F^0[T^*(T, H), H_s^*(H_s, H)] , \tag{34}$$

where

$$T^*(T, H) = \frac{T_N}{T_N(H)} T , \quad H_s^*(H_s, H) = \frac{T_N}{T_N(H)} H_s . \tag{35}$$

T_N is the transition temperature in zero field, $T_N = T_N(H = 0)$, and $F^0(T^*, H_s^*)$ represents the droplet formula (5) for an antiferromagnet at temperature T^* and staggered field H_s^* and in zero external magnetic field. From (34) we deduce that in our approximation the effect of the magnetic field on the antiferromagnetic transition is simply described by a renormalization of the variables T and H_s. This behavior coincides with that assumed by Fisher[14] in his theory of "renormalization" and, in particular, is found to hold true for a certain decorated Ising model[16] in an external field. Because T^* and H_s^* are the simple linear functions (35) of T and H_s, the critical exponents for the order parameter M_s, the staggered susceptibility χ_s, the specific heat C_H, and the critical isotherm for fixed H coincide with the values in zero field, so that they are,[9] respectively,

$$\beta_1 = \beta = \frac{\tau_1 - 2}{\sigma_1} , \qquad \gamma_1 = \gamma_1' = \gamma = \frac{3 - \tau_1}{\sigma_1} ,$$
$$\alpha_1 = \alpha_1' = \alpha = 2 - \frac{\tau_1 - 1}{\sigma_1} , \quad \delta_1 = \delta = \frac{1}{\tau_1 - 2} , \tag{36}$$

where the usual notation for the critical exponents[7] has been used and the index 1 indicates that the exponents refer to the antiferromagnetic transition in a finite field H. The critical behavior at $T_N(H)$ of the magnetization M and of the susceptibility χ is related to the behavior of the entropy S and of the specific heat C_H. In fact, from (34) and (35) one finds near $T_N(H)$

$$M(T, H) \sim -T_N'(H) S(T, H) ,$$
$$\chi(T, H) \sim T_N^{-1}(H) [T_N'(H)]^2 C_H(T, H) - T_N''(H) S(T, H) , \tag{37}$$

where T_N' and T_N'' denote the first and the second field derivatives of $T_N(H)$. Relations (37) refer to the most singular parts of the different quantities. From (37) and (34) we deduce, if $H \neq 0$,

$$\Delta M \equiv M(T, H) - M(T_N(H), H) \sim |\epsilon_1|^{1-\alpha} ,$$
$$\chi(T, H) \sim |\epsilon_1|^{-\alpha} , \tag{38}$$
$$\epsilon_1 = (T_N(H) - T)/T_N(H) . \tag{39}$$

In zero field $T_N' = 0$ and $T_N'' \neq 0$ [see Eq. (19)] so that the susceptibility remains finite and its exponent is $(1 - \alpha)$, in agreement with previous results.[17] We have assumed that the constants appearing in (32) (for example, q_0 and B_1) are temperature independent. Should these constants have an analytic temperature dependence, (34) would not apply. However, (36) and (38) continue to hold true in this case also, but one finds that the relative amplitudes are temperature dependent.

If $H = H_t$, so that $T_N(H_t) = T_t = T_m(H_t)$ and the variable Y_2 defined in (16) approaches the value unity, the terms F_2 and G given by (14) and (33) are also nonanalytic at T_N. This nonanalyticity corresponds to the essential singularity that the usual droplet formula (5) has at $Y_1 = 1$. However, if $X \neq 1$, this singularity is extremely weak because all the derivatives remain finite but the radius of convergence of the Taylor series is zero.[9] The same kind of singularity is found for F_2 when $Y_2 = 1$ and for G when $Y_2 = 1$ and $Y_1 = 1$. It is clear, however, that these weak singularities superimposed to the stronger singularity of F_{1s} play no role in the determination of the critical exponents. We conclude that, if the tricritical point is approached with constant H, the relative critical exponents, denoted by an index t, are the same as those relative to the antiferromagnetic transition:

$$\beta_t = \beta_1 , \ \gamma_t = \gamma_1 , \ \alpha_t = \alpha_1 , \ \delta_t = \delta_1 . \tag{40}$$

These same exponents, but now relative to the appropriate variables, also apply if the tricritical point is approached in the $H_s = 0$ plane along any other line which is not asymptotically parallel to $T_N(H)$ and $T_m(H)$. For example, if the (H_t, T_t) point is approached keeping the temperature fixed, the exponents in (40) are relative to the variable $(H_t - H)/H_t$ in place of ϵ_1.

In case the tricritical point is approached along the metamagnetic line $T_m(H)$ we have to specify, in the first place, which kind of contact the two curves $T_N(H)$ and $T_m(H)$ have at the crossing point (H_t, T_t). In the mean-field theory[5] the two curves have a

common tangent and different second derivatives. In the present model, the two curves do not have any particular relation at the tricritical point so that it seems natural that the two curves should have a different tangent. However, some form of interaction, which we have not considered, between the different clusters might change the situation. So, to be more general, we assume that

$$T_N(H) - T_m(H) \sim G(H - H_t)^g \tag{41}$$

for $H > H_t$, where g is an integer. Measurements in some metamagnets suggest that the first derivative of the phase boundary[1-3] is continuous at (H_t, T_t). However, the measurements are not yet able to determine with great precision the phase boundary in particular near the tricritical point, so that a small discontinuity cannot be excluded.

Along the $T_m(H)$ curve, the basic variables [(15), (16)] take the following values near the tricritical point:

$$\begin{aligned} &Y_1 = 1 , \\ &X_1 = \exp\left(-a_1 \frac{T_N(H) - T_m(H)}{T_m(H)}\right) \\ &\quad \sim \exp\left(-a_1 \frac{G}{T_t}(H - H_t)^g\right) \\ &\quad \sim \exp\left[-a_1 \frac{G}{T_t}\left(\frac{T_t \epsilon_m}{T'_{mt}}\right)^g\right] , \end{aligned} \tag{42}$$

$$Y_2 = 1 , \quad X_2 \sim \bar{X}_2 = \exp[-a_2(T^* - T_t)/T_t] ,$$

$$\epsilon_m = [T_m(H) - T_t]/T_t , \tag{43}$$

$$a_i = \bar{a}_i \bar{\omega}_i / K_B \quad (i = 1, 2) , \tag{44}$$

where $T'_m = [dT_m(H)/dH]_{H_t}$ and we have taken into account (41). The contribution to the magnetization which comes from (32) is similar to the contribution relative to the magnetization near $T_N(H)$, with the only difference being that $\ln X_1 \sim \epsilon_m^g$ in place of $\ln X_1 \sim \epsilon_1$. We conclude that M has a singular term proportional to $(-\epsilon_m)^{g(1-\alpha)}$, where α is the specific-heat exponent [see (36)]. The contribution to M from (14) is

$$\begin{aligned} (K_B T)^2 q_2 \sum_{l_2=1}^{\infty} & l_2^{-\tau_2} Y_2^{l_2} X_2^{\lambda_2(l_2)} \left(l_2 \frac{\partial \ln Y_2}{\partial H} f_{21}(l_2) f_{22}(l_2)\right. \\ & - \frac{a_1}{T} T'_N(H) B_{21} (B_{21} X_1^{b_{21}\lambda_1(l_2)} Y_1^{l_2} + 1)^{-2} \\ & \left. \times b_{21} l_2^{g_1} X_1^{b_{21}\lambda_1(l_2)} Y_1^{l_2} f_{22}(l_2)\right) , \end{aligned} \tag{45}$$

and it is well behaved near (H_t, T_t) because $X_2 \sim \bar{X}_2 < 1$. This expression, in particular the first term, is similar to the expression for the magnetization of a ferromagnet in zero external field at a temperature below its critical temperature. However, a profound difference from this case is due to the presence of the factor f_{21} in (45). As was discussed in Sec. II, the first-order phase transition terminates at (H_t, T_t) because when $X_1 > 1$, i.e., for $T_t < T < T^*$, the fluctuations due to F clusters are depressed by f_{21}, and at T^*, in particular, there is no singularity. Adding together (45), which has a linear behavior in the variable ϵ_m, with the singular term coming from F_1, the magnetization along $T_m(H)$ near (H_t, T_t) reads

$$M_-[T_m(H)] \sim M_t + L\epsilon_m - P|\epsilon_m|^{g(1-\alpha)} , \tag{46}$$

where M_t is the value of the magnetization at the tricritical point and L and P are positive constants. If the $T_m(H)$ line is approached from the paramagnetic phase, the magnetization can be obtained by using the appropriate expression (24) for the free energy. A calculation similar to the previous one gives

$$M_+[T_m(H)] \sim M_t - \bar{L}\epsilon_m - \bar{P}|\epsilon_m|^{g(1-\alpha)} , \tag{47}$$

where $\bar{L}$ and $\bar{P}$ are positive constants and the (M, T) phase diagram has the shape shown in Fig. 5 in the case $g > 1$. If $g = 1$, $M_\pm$ approach M_t with a vertical tangent corresponding to the $|\epsilon_m|^{1-\alpha}$ term in (46) and (47). This term, however, might be difficult to observe experimentally because this singular part is strictly related to the singular part of the specific heat in zero field and it is well known that the singularity in the specific heat is extremely weak when compared to the singularities of other quantities. We notice that the coefficients L and $\bar{L}$ in (46) and (47) become larger the closer T^* gets to T_t. For the susceptibility along $T_m(H)$ one finds that F_2 gives a contribution which is regular whereas F_1 gives

$$\chi_- \sim (-\epsilon_m)^{-g\alpha} . \tag{48}$$

As in all other cases, $\alpha = 0$ implies a logarithmic behavior. The same behavior is found for the paramagnetic phase at the metamagnetic transition

$$\chi_+ \sim (-\epsilon_m)^{-g\alpha} . \tag{49}$$

The (M, T) phase diagram we have obtained is similar to the one obtained on the basis of "classical" theories.[8] There are two basic differences. In the first place, M_+ need not be the continuation of the magnetization $M[T_N(H)]$ along the antiferromagnetic transition because the free energy is not analytic at (H_t, T_t) and Landau's argument[6] does not apply. In fact, we do not expect this behavior because the slope of M_+ depends on how large $(T_t - T^*)/T_t$ is, whereas the slope of $M[T_N(H)]$ basically depends on the field dependence of the surface energy of AF clusters. In the second place, M also has a nonanalytic part and the sus-

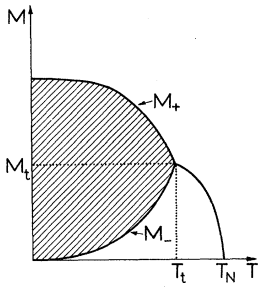

FIG. 5. Schematic M, T phase diagram for a metamagnet of first kind. The hatched area represents a region of coexisting phases.

ceptibility is divergent at $(H_t,\ T_t)$ along both M_+ and M_-. The mean-field theory predicts a finite susceptibility when $(H_t,\ T_t)$ is approached along M_+ and a divergent one for M_-. Experiments in liquid He^3-He^4 mixtures show[13] that the phase separation diagram is similar to Fig. 5 if M is interpreted as the He^3 concentration x. It is found that x_+ is not the continuation of x along the λ line, and the quantity corresponding to the susceptibility is divergent[8] along x_+. As was discussed before, it is not yet certain if the phase diagram of metamagnets has the shape given in Fig. 5 or not. In any case, experiments in DAG[3] clearly show that if M_+ linearly approaches M_t, then M_+ is not the continuation of $M[T_N(H)]$ and χ_+ is divergent at the tricritical point.

The free energy and the equation of state near the tricritical point does not have the simple homogeneous form that has been suggested by Griffiths.[8] In fact, in the region of the $(H,\ T)$ plane below the $T_m(H)$ line, i.e., $T < T_m(H)$ for given H, the most singular contribution to the free energy comes from the F_{1s} term [Eq. (32)]. Taking into account (34) and (35) and that[7] $F^0(T^*,\ 0) \sim A(|T^* - T_N|/T_N)^{2-\alpha}$, F_{1s} reads

$$F_{1s}(T_t + \Delta T,\ H_t + \Delta H,\ 0) \sim |\Delta T|^{2-\alpha}\phi_{1-}(\Delta H/\Delta T) + F_{1s}(T_t,\ H_t,\ 0), \quad (50)$$

where $\phi_{1-}(x)$ is a suitable function. The contributions (33) and (13) to F can be represented by the first few terms of their expansions in power of ΔT and ΔH [these are asymptotic expansions due to the essential singularity that (33) and (13) have at the tricritical point]. We conclude that the deviation of the magnetization from its value at $(H_t,\ T_t)$ can be written in the form

$$\Delta M = M - M_t \sim \frac{\Delta T}{|\Delta T|}\,|\Delta T|^{1-\alpha}\,\psi_{1-}\left(\frac{\Delta H}{\Delta T}\right) + \Delta T\,\psi_{2-}\left(\frac{\Delta H}{\Delta T}\right), \quad (51)$$

where $\psi_{1-}(x)$ and $\psi_{2-}(x)$ are two suitable functions which can be explicitly obtained from (12) and (13). We notice that $\psi_{1-}(x)$ has a zero of order $(1-\alpha)$ at $x = x_t$, where $x_t = [\partial T_N(H)/\partial H]^{-1}_{H_t}$, so that, taking into account (41), Eq. (46) results when the tricritical point is approached along $T_m(H)$.

For the region of the $(H,\ T)$ plane above the $T_m(H)$ line, i.e., $T > T_m(H)$, by using (24)–(26) for the free energy, an expression similar to (51) is obtained with two functions $\psi_{1+}(x)$ and $\psi_{2+}(x)$. The choice of the constants appearing in F and $\tilde{F}$, as discussed at the end of Sec. II, gives a constraint on $\phi_{1\pm}$ and $\phi_{2\pm}$, so that M is continuous across $T_m(H)$ for $H < H_t$. Moreover, (46) and (47) guarantee that $\psi_{2-}(x_t) > 0$ and $\psi_{2+}(x_t) < 0$, in agreement with Fig. 5.

A straightforward calculation gives the staggered magnetization M_s and susceptibility χ_s and the specific heat at constant fields H and H_s. The only singular contributions arise from those terms relative to the field or temperature derivatives of Y_1 and X_1 because the derivatives of Y_2 and X_2 bring a factor $X_2^{\lambda_2(i_2)}$ which gives convergence. This is why the singular parts for these quantities have the same form as in zero field and we find

$$M_s \sim (-\epsilon_m)^{\beta g},\quad \chi_s \sim |\epsilon_m|^{-\gamma g}, \quad (52)$$

$$C_H \sim |\epsilon_m|^{-\alpha g}, \quad (53)$$

where β, γ, and α are the zero-field exponents (36). The exponents (52) differ from (36) for the factor g. This is due to the form (42) for X_1, which takes into account the fact that, if $g > 1$, the AF singularity is approached along a curve asymptotically tangent to $T_N(H)$.

Up to this point it has been assumed that $T_N(H)$ and $T_m(H)$ are analytic functions of H also at H_t, so that (41) implies an AF phase boundary with a continuous first derivative at $(H_t,\ T_t)$ if $g > 1$. However, (41) for $H > H_t$ represents an effective temperature distance for AF clusters from their seeming critical point and we cannot exclude that this effective distance is not related to the shape of the phase boundary because $g > 1$ implies some form of interaction between clusters. If this is true, then $T_N(H)$ might be nonanalytic at H_t; for instance, $T_N(H)$ could have a discontinuity at H_t in its first derivative. This has no direct observable consequences because $T_N(H)$ for $H > H_t$ does not represent a phase boundary but it serves only to give a measure of the surface energy of AF clusters along $T_m(H)$. In this case, g [in (41)] must be interpreted as a new exponent, unrelated to the shape of the antiferromagnetic phase boundary. Precise measurements of the phase boundary and of M_s along $T_m(H)$ and for $H = H_t$ should be very valuable in clarifying this point.

V. METAMAGNET OF SECOND KIND

In this section we consider the case in which both AF and F clusters become critical at the tricritical

point, i.e., $T^* = T_t$. It is obvious that the critical behavior along the antiferromagnetic line $T_N(H)$ for $H < H_t$ is exactly the same as in the case $T^* > T_t$ considered in Sec. IV. In fact, (36) and (38) still apply because the F_2 term of the free energy [Eq. (12)] is regular on this line as in the previous case and the only singular contribution comes from the F_{1s} term [Eq. (32)] which depends only on the variables relating to the AF clusters.

In studying the behavior near the tricritical point, approached from the antiferromagnetic phase, it is useful to split the free energy as in Sec. IV as follows:

$$F(T, H, 0) = -U_0 + F_{1s}(T, H, 0) + G(T, H, 0) + F_2(T, H, 0) , \quad (54)$$

where these three parts are given by (32), (33), and (13), respectively. We begin by studying the critical behavior along the $T_m(H)$ line. The three basic variables Y_1, X_1, and Y_2 take the values given in (42), whereas X_2 is

$$X_2 = \exp(a_2 \epsilon_m) , \quad (55)$$

where a_2 is given in (44) and ϵ_m is the reduced temperature difference [(43)]. Since F_{1s} depends only on Y_1 and X_1, the singular part of this term is the same as we have calculated in Sec. IV. In particular, it gives a term proportional to $|\epsilon_m|^{g(1-\alpha)}$ for the magnetization. In the present case, however, G and F_2 also contain a singular part because now $X_2 = 1$ at the tricritical point. In the first place we consider the contribution [(45)] to M which comes from F_2. To obtain the singular contribution, the sum over l_2 can be approximated by an integral[9] so that the first term of (45) is proportional to

$$\mathcal{T} = \int_1^\infty d l_2 \, l_2^{1-\tau_2} [\exp(\epsilon_m l_2^{\sigma_2}) - 1] \sim |\epsilon_m|^{(\tau_2 - 2)/\sigma_2} , \quad (56)$$

because the factors f_{21} and f_{22} play an inessential role in this case ($\epsilon_m < 0$). The second term of (45) is proportional to the integral

$$\mathcal{J} = \int_1^\infty d l_2 \, l_2^{\sigma_1 - \tau_2} [\exp(\epsilon_m l_2^{\sigma_2} - |\epsilon_m|^g l_2^{\sigma_1}) - 1] . \quad (57)$$

This integral has a different asymptotic behavior for $\epsilon_m \to 0$ depending on the values of g and of σ_2/σ_1. If $g = 1$ and $\sigma_2/\sigma_1 < 1$, one obtains

$$\mathcal{J} \sim |\epsilon_m|^{(\tau_2 - 1)/\sigma_1 - 1} ; \quad (58)$$

otherwise

$$\mathcal{J} \sim |\epsilon_m|^{(\tau_2 - 1 - \sigma_1)/\sigma_2} , \quad (59)$$

but this term is always negligible with respect to (56) because $\sigma_1 < 1$. One can easily calculate $\partial G/\partial H$ from (33) to obtain the relative contribution to the magnetization. A first term comes from the field derivative of X_1, and the singular part is proportional to

$$\mathcal{J}' = \int_1^\infty d l_1 \, l_1^{\sigma_1 - \tau_1} [\exp(-|\epsilon_m|^g l_1^{\sigma_1}) - 1] \sim |\epsilon_m|^{g(1-\alpha)} , \quad (60)$$

where α is given in (36). A second term comes from the derivative of Y_2 contained in f_{12} and the singular part is proportional to

$$\mathcal{T}' = \int_1^\infty d l_1 \, l_1^{1-\tau_1} [\exp(-|\epsilon_m|^g l_1^{\sigma_1} + \epsilon_m l_1^{\sigma_2}) - 1] \sim |\epsilon_m|^{\min[(\tau_1 - 2)/\sigma_2 , \, g(\tau_1 - 2)/\sigma_1]} , \quad (61)$$

where, in writing the asymptotic behavior, we have assumed that $\sigma_1/\sigma_2 \leq 2$. Collecting together all the different terms we obtain finally

$$M_- - M_t \sim |\epsilon_m|^\zeta , \quad (62)$$

$$\zeta = \min \left[\frac{\tau_2 - 2}{\sigma_2} , \, \frac{\tau_1 - 2}{\sigma_2} , \, g \, \frac{\tau_1 - 2}{\sigma_1} , \, g(1 - \alpha) , \, g \, \frac{\tau_2 - 1 - \sigma_1}{\sigma_1} \right] , \quad (63)$$

where the last term of (63) has to be taken into account only if $\sigma_2 < \sigma_1$.

If $g > 1$ the last three terms of (63) are certainly negligible with respect to the first two, g being an integer. Also, if $g = 1$ it seems very likely that the last two terms of (63) are negligible because we expect that σ_2 and τ_2 are close to σ_1 and τ_1 ($\sigma_1 = \frac{16}{25}$, $\tau_1 = 2\frac{1}{5}$ for the $3D$ Ising model) or close to the $2D$ Ising model values ($\sigma = \frac{8}{15}$, $\tau = 2\frac{1}{15}$) if the F clusters are two dimensional in character. Then the two last terms of (63) are close to unity and therefore larger than the remaining terms. We also note that the amplitudes relative to the different terms of M are expected to be rather different in magnitude, the one relative to (56) being the largest unless the interference effects between clusters is very large. In fact, this contribution is due to the direct effect of fluctuations of ferromagnetic kind on the magnetization, whereas the other terms are due to interference effects between different clusters and thus they can be considered as a second-order effect. Therefore, even if the asymptotic behavior is given by (63) we may expect that, in any case, the dominant behavior in the experimentally accessible region of ϵ_m is characterized by the exponent $\zeta = (\tau_2 - 2)/\sigma_2$.

With a very similar analysis it is possible to calculate the asymptotic behavior of the different contributions to the susceptibility χ and to the specific heat C_H in constant magnetic field. If we define two new exponents θ and α_2 for χ and C_H by

$$\chi \sim |\epsilon_m|^{-\theta} , \quad C_H \sim |\epsilon_m|^{-\alpha_2} , \quad (64)$$

we find

$$\theta = \alpha_2 = \max\left[\frac{3-\tau_2}{\sigma_2}\,,\ \min\left(\frac{3-\tau_1}{\sigma_2}\,,\ g\,\frac{3-\tau_1}{\sigma_1}\right)\right]\,, \tag{65}$$

where we have neglected terms of order α on the ground of the discussion following (63). For M_s and χ_s along $T_m(H)$ we find

$$\beta_2 = \min\left(g\,\frac{\tau_1-2}{\sigma_1}\,,\ \frac{\tau_2-2}{\sigma_2}\,,\ \frac{\tau_1-2}{\sigma_2}\,,\ g\,\frac{\tau_2-2}{\sigma_1}\right)\,, \tag{66}$$

$$\gamma_2 = \max\left(g\,\frac{3-\tau_1}{\sigma_1}\,,\ \frac{3-\tau_2}{\sigma_2}\right)\,, \tag{67}$$

having defined β_2 and γ_2 by $M_s \sim |\epsilon_m|^{\beta_2}$ and $\chi_s \sim |\epsilon_m|^{-\gamma_2}$. In this case the amplitudes of the terms with exponents $g(\tau_1-2)/\sigma_1$ and $(\tau_2-2)/\sigma_2$ in (66), as well as both terms of (67), are expected to be of comparable size because both variables Y_1 and Y_2 have a nonzero derivative with respect to H_s. The other terms of (66) are due to interference effects.

By using (24)–(26), the same exponents (63) and (65) are also found when the tricritical point is approached along $T_m(H)$ in the paramagnetic phase; only the relative amplitudes are different. For the magnetization $M_+ - M_t$, in particular, the amplitude of the term with exponent $(\tau_2-2)/\sigma_2$ is negative, whereas the similar amplitude for $M_- - M_t$ is positive, thus corresponding to the behavior given in Fig. 6. This difference in sign is due to the fact that in (25) the cluster contribution to the free energy is proportional to $Y_2^{-\tau_2}$ while in (14) it is proportional to $Y_2^{\tau_2}$.

One might be interested in the critical behavior when the tricritical point is approached from the AF phase along a direction different from the one of $T_m(H)$. For instance, if we keep the magnetic field fixed ($H = H_t$), then the basic variables [(6), (15), (16)] become

$$Y_1 = 1\,,\quad \ln X_1 \sim \epsilon_2\,,\quad \ln Y_2 \sim \epsilon_2\,,\quad \ln X_2 \sim \epsilon_2\,,$$
$$\epsilon_2 = (T - T_t)/T_t\,, \tag{68}$$

and the calculations proceed in a way similar to the previous case. The main difference is that the singular contributions to the different quantities are proportional to integrals similar to (56) or (57), with an extra factor $\exp(\epsilon_2 l)$ wherever the variable Y_2 is involved. We find

$$M - M_t \sim |\epsilon_2|^{\min(\tau_1-2,\,\tau_2-2)}\,, \tag{69}$$

$$\chi \sim C_H \sim |\epsilon_2|^{-\max(3-\tau_1,\,3-\tau_2)}\,. \tag{70}$$

These same exponents apply for any other direction which is not asymptotically parallel to $T_m(H)$ or to $T_N(H)$ at $(H_t,\ T_t)$.

We now study whether or not homogeneity in the $(H,\ T)$ plane applies in the case of a metamagnet of the second kind near its tricritical point. In general, it is not possible to write the free energy in a homogeneous form because of the different exponents which enter the different terms of F. However, if the interference effects, i.e., B_{12} and B_{21} in (18), are small, M, χ, and C_H should be dominated by the ferromagnetic fluctuations so that the only important term of F in the $H_s = 0$ plane is given by (13), with $f_{21} = 1$. For $T = T_t(1+\epsilon_2)$ and $H = H_t + \Delta H$, and taking into account (15), (16), and (20)–(22), this term reads

$$-q_2 K_B T_t \sum_{l_2=1}^{\infty} l_2^{-\tau_2} \exp(-dv\,l_2 + a_2\,\epsilon_2\,l_2^{\sigma_2}) \times [B_2 \exp(a_2\,b_2\,\epsilon_2\,l_2^{\sigma_2}) + 1]^{-1}\,, \tag{71}$$

where $d = -K_B^{-1}(\partial E_2/\partial T)_{H_t,T_t}$, a_2 is given in (44), and

$$v = (T'_{mt}\,\Delta H - T_t\,\epsilon_2)/T_t \tag{72}$$

is a measure of the distance from the $T_m(H)$ line and it is non-negative in the AF phase. T'_{mt} is the field derivative of $T_m(H)$ at $(H_t,\ T_t)$. The contribution to the magnetization which comes from (71) can be written in the form

$$\Delta^* M = v^{\tau_2-2}\,g_-(\epsilon_2/v^{\sigma_2})\,, \tag{73}$$

$$g_-(z) = R\int_0^{\infty} dx\,x^{1-\tau_2}\{(1+B_2)^{-1} - \exp(-x + hz\,x^{\sigma_2}) \times [B_2 \exp(b_2\,hz\,x^{\sigma_2}) + 1]^{-1}\}\,, \tag{74}$$

where R and h are postive constants and we have again approximated the series (71) by an integral. If we are interested in $\Delta^* M$ near the $T_m(H)$ line, it is more useful to write (73) in the form

$$\Delta^* M = |\epsilon_2|^{(\tau_2-2)/\sigma_2}\,h_-(\epsilon_2/v^{\sigma_2})\,, \tag{75}$$

$$h_-(z) = R'\int_0^{\infty} dy\,y^{(2-\tau_2-\sigma_2)/\sigma_2} \times\left\{(1+B_2)^{-1} - \exp\left[-h'\left(\frac{y}{|z|}\right)^{\sigma_2^{-1}} + \frac{z}{|z|}\,y\right] \times\left[B_2 \exp\left(b_2\,\frac{z}{|z|}\,y\right) + 1\right]^{-1}\right\}\,, \tag{76}$$

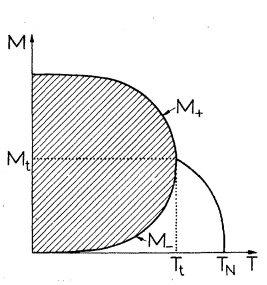

FIG. 6. As for Fig. 5 for a metamagnet of second kind.

where R' and h' are two other constants. The condition that M is continuous across $T_m(H)$ for $T > T_t$ implies that B_2 and b_2 are such that $h_-(+\infty) = 0$. In this case one finds

$$h_-(z) \underset{z \to \infty}{\to} |z|^{-\sigma_2^{-1}} . \tag{77}$$

If $v < 0$ the system is in the paramagnetic phase and (25) must be used. An expression similar to (73) is obtained:

$$\Delta^* M = -|v|^{\tau_2 - 2} g_+(\epsilon_2/|v|^{\sigma_2}) , \tag{78}$$

where $g_+(z)$ has an expression similar to (74) but with different constants. In (73) and (78) the asterisk indicates that, in general, these terms do not give the asymptotically correct singular part of the magnetization but they only give the contribution of the ferromagnetic clusters which should give the dominant part. In this case the exponents introduced in (62) and (64) have the values

$$\zeta^* = (\tau_2 - 2)/\sigma_2 , \quad \theta^* = \alpha_2^* = (3 - \tau_2)/\sigma_2 , \tag{79}$$

and in place of (69) and (70) we have

$$\Delta^* M \sim |\epsilon_2|^{\tau_2 - 2} , \quad \chi^* \sim C_H^* \sim |\epsilon_2|^{-(3-\tau_2)} . \tag{80}$$

The homogeneous forms (73) and (78), but with different functions $g_\pm(z)$ and the exponents (79) and (80), are exact in the case $\sigma_1 = \sigma_2$, $\tau_1 = \tau_2$, and $g > 1$ in (41). In this case, taking into account (36), the following relations exist:

$$\zeta = \beta , \quad \theta = \alpha_2 = \gamma .$$

The homogeneous forms (73) and (78) are equivalent to the homogeneous form for an ordinary critical point, with M playing the role of the order parameter and v the role of the field coupled to the order parameter, if one takes into account that σ_2 and $(\tau_2 - 2)$ correspond to $(\gamma + \beta)^{-1}$ and δ^{-1}, respectively, with the usual notation for the critical indices.[7] The homogeneous form for the free energy near a tricritical point proposed by Griffiths[8] corresponds to a particular case of (73), and (78) when

$$\sigma_2 = \tau_2 - 2 = \mu , \tag{81}$$

where μ is the index introduced by Griffiths. In this case the exponents (80) turn out to be $\zeta = 1$ and $\theta = \mu^{-1} - 1$, thus corresponding to a phase diagram as given in Fig. 5. The present picture of a tricritical point is rather different from the one given by Griffiths. In our case, in fact, we find the linear behavior for $M_\pm$ in the case $T^* > T_t$, but then the divergence of χ and C_H are due to the antiferromagnetic singularity, which is absent in the Griffiths case, and the free energy cannot be represented by a homogeneous function. This representation is possible in the case $T^* = T_t$, but for the present model the relation $\sigma_2 = \tau_2 - 2$, which is necessary to obtain $\zeta = 1$, seems artificial, even if it cannot be excluded in principle. On the other hand, even if this relation applies, it is very likely that the values (79) for the exponents do not give the true asymptotic behavior. For instance, using the $3D$ Ising model values for σ_1 and τ_1, both $(\tau_1 - 2)/\sigma_2$ for $\sigma_2 > \frac{1}{5}$ and $g(\tau_1 - 2)/\sigma_1$, if $g = 1$ or 2, turn out to be smaller than unity so that $\zeta = 1$ is not given by (63) and ζ more likely lies in the range $\frac{1}{3} - \frac{2}{3}$. However, in this case too, the values (79) might describe the dominant part of the critical behavior for most of the range of ϵ_2, but this implies that the interference effects must be very small; the larger the difference between $\zeta = 1$ and the value given by (63), the smaller the interference effects must be.

VI. RENORMALIZED CRITICAL BEHAVIOR

In Secs. IV and V it has been assumed that the local magnetic field H is under control. In practice, the experiments are performed in fixed external field $H_0 = H + NM$, where N is the demagnetizing factor of the sample, so that the observed critical behavior is renormalized.[14] In the magnetic case it is experimentally feasible to express the different quantities in terms of the local field, so that the unrenormalized critical behavior can be obtained. However, it is useful to know the theoretical renormalized behavior, both because the needed experimental data are not always available and because in the reduction process one may lose accuracy. At the antiferromagnetic transition $T_N(H)$ the critical behavior is renormalized according to Fisher's theory.[14] In fact, the form of the singular part (34) of the free energy has exactly the form considered by Fisher and we conclude that the exponents (36) become, respectively,

$$\begin{aligned} &\beta_{1x} = \beta/(1 - \alpha) , \quad \gamma_{1x} = \gamma/(1 - \alpha) , \\ &\delta_{1x} = \delta/(1 - \alpha) , \quad \alpha_{1x} = -\alpha/(1 - \alpha) . \end{aligned} \tag{82}$$

In a similar way the renormalized behavior of M and χ can be obtained by the use of (37) and it turns out to be

$$\Delta M \sim |\epsilon_1|^{(1-\alpha)^{-1}} , \quad \Delta\chi \sim |\epsilon_1|^{\alpha/(1-\alpha)} . \tag{83}$$

The renormalization [Eqs. (82) and (83)] also holds true at T_t in the case of a metamagnet of the first kind. In fact, as has been discussed in Sec. IV, F clusters introduce a term which has only an essential singularity at T_t, with all the derivatives remaining finite. For our purpose this term can be assimilated to a regular function and Fisher's results still apply. Expressions (46) and (47) for the magnetization at the metamagnetic transition are not changed by renormalization because the phase boundary is uniquely determined, independently of the constraint one imposes.

To obtain the renormalized critical behavior of a metamagnet of the second kind it is necessary to take into account also the ferromagnetic singularity, and we cannot make use of Fisher's results. In the first place, for the specific heat we have to take into account the relation[18]

$$C_{H_0} = C_H - T\left(\frac{\partial M}{\partial T}\right)_H^2 \left[\frac{1}{N} + \left(\frac{\partial M}{\partial H}\right)_T\right]^{-1} \tag{84}$$

between the specific heat in constant external field C_{H_0} and the specific heat in constant local field C_H. The second term on the right-hand side of (84) cancels the strong divergence of C_H as given by (65) or (70), depending on the direction of approach to (H_t, T_t), and leaves a much weaker divergence. We find

$$C_{H_0} \sim A_1 (T'_{Nt} - T'_{mt})^2 |\epsilon_2|^{-c_1\alpha} + A_2 |\epsilon_2|^{-\bar{\alpha}} , \tag{85}$$

where A_1 and A_2 are constants, T'_{Nt} and T'_{mt} indicate the field derivatives of $T_N(H)$ and $T_m(H)$ at (H_t, T_t), and α is given in (36). If (H_t, T_t) is approached along $T_m(H)$, then c_1 and $\bar{\alpha}$ read

$$c_1 = g ,$$
$$\bar{\alpha} = \max\left(\sigma_2^{-1}[1 + 2\sigma_2 - \min(\tau_1, \tau_2)] , g\alpha - \frac{1-\sigma_2}{\sigma_2}\right), \tag{86}$$

otherwise, c_1 and $\bar{\alpha}$ read

$$c_1 = 1 , \quad \bar{\alpha} = 1 + 2\sigma_2 - \min(\tau_1, \tau_2) . \tag{87}$$

Since $\bar{\alpha}$ is of the order of α, we conclude that the specific heat in constant external field at (H_t, T_t) has a divergence with an exponent similar to the one of the specific heat for $H = 0$. The calculation of (85) is straightforward but rather lengthy. We proceed in the following way. The free energy is considered as a function of the variables X_1, X_2, Y_2 which were defined in (15) and (16) ($H_s = 0$ so $Y_1 = 1$): $F(T, H, 0) = \mathcal{F}(X_1, X_2, Y_2)$. Taking into account (19), (22), and (44), at the tricritical point the field and temperature derivatives of these variables are

$$\left(\frac{\partial X_1}{\partial T}\right)_t = \frac{a_1}{T_t} , \quad \left(\frac{\partial X_1}{\partial H}\right)_t = -\frac{T'_{Nt} a_1}{T_t} ,$$
$$\left(\frac{\partial X_2}{\partial T}\right)_t = \frac{a_2}{T_t} , \tag{88}$$

$$\left(\frac{\partial Y_2}{\partial T}\right)_t = \frac{d}{T_t} , \quad \left(\frac{\partial Y_2}{\partial H}\right)_t = -\frac{T'_{mt} d}{T_t} , \tag{89}$$

where d is defined following Eq. (71). Then we obtain

$$C_H = -T\left(\frac{\partial^2 F}{\partial T^2}\right)$$
$$= -T_t^{-1}(a_1^2 \mathcal{F}_{X_1X_1} + d^2 \mathcal{F}_{Y_2Y_2} + a_2^2 \mathcal{F}_{X_2X_2}$$
$$+ 2a_1 d \mathcal{F}_{X_1Y_2} + 2a_1 a_2 \mathcal{F}_{X_1X_2}$$
$$+ 2a_2 d \mathcal{F}_{X_2Y_2} + \cdots) , \tag{90}$$

where $\mathcal{F}_{XY}$ indicates the second derivative of $\mathcal{F}$ with respect to the displayed arguments, and the dots in (90) indicate terms containing the second derivatives of the variables X_1, X_2, Y_2 and terms relative to the deviation of the first derivatives from their values [Eqs. (88) and (89)] at the tricritical point. It is possible to show that those terms that are not explicitly written in (90) give a negligible contribution. Expressions similar to (90) can be obtained for $\partial M/\partial H = -\partial^2 F/\partial H^2$ and $\partial M/\partial T = -\partial^2 F/\partial H \partial T$, and we do not write them explicitly. Use of these expressions and of (90) in (84) gives (85) if we take into account the following asymptotic behavior of the second derivatives of $\mathcal{F}$, which can be obtained by proceeding as in Sec. V:

$$\mathcal{F}_{Y_2Y_2} \sim |\epsilon_2|^{c_2(\tau_2-3)} + |\epsilon_2|^{c_3(\tau_1-3)} ; \tag{91}$$

$$\mathcal{F}_{X_1Y_2} \sim |\epsilon_2|^{c_3(\tau_1-2-\sigma_1)} + |\epsilon_2|^{c_3(\tau_2-2-\sigma_1)} ,$$
$$\mathcal{F}_{Y_2X_2} \sim |\epsilon_2|^{c_3(\tau_1-2-\sigma_2)} + |\epsilon_2|^{c_2(\tau_2-2-\sigma_2)} ; \tag{92}$$

$$\mathcal{F}_{X_1X_1} \sim |\epsilon_2|^{c_3(\tau_2-2\sigma_1-1)} + |\epsilon_2|^{c_1(\tau_1-2\sigma_1-1)/\sigma_1} ,$$
$$\mathcal{F}_{X_2X_2} \sim |\epsilon_2|^{c_3(\tau_1-2\sigma_2-1)} + |\epsilon_2|^{c_2(\tau_2-2\sigma_2-1)} ,$$
$$\mathcal{F}_{X_1X_2} \sim |\epsilon_2|^{c_3(\tau_1-1-\sigma_1-\sigma_2)} + |\epsilon_2|^{c_3(\tau_2-1-\sigma_1-\sigma_2)} . \tag{93}$$

Here the constants c_i take the values $c_1 = g$, $c_2 = \sigma_2^{-1}$, $c_3 = \min(\sigma_2^{-1}, g\sigma_1^{-1})$, if (H_t, T_t) is approached along $T_m(H)$, otherwise they take the value unity. In (91) and (93) we have not written, for simplicity, the coefficients in front of each temperature-dependent term.

Having obtained the singular part (85) for C_{H_0}, we are now in a position to study the renormalized behavior of C_{H_0}, M, and χ when the tricritical point is approached, keeping the external field H_0 fixed to the critical value $H_{0t} = H_t + NM_t$. For simplicity, we consider only the case $\sigma_1 = \sigma_2$ and $\tau_1 = \tau_2$ or the case in which the ferromagnetic clusters dominate the fluctuations near the tricritical point so that we need to consider only the term (13) with $f_{21} = 1$. In this case we can use (73) for M and the constraint equation $H_{0t} = H + NM$ takes the form

$$\Delta H = N v^{(\tau_2-2)/\sigma_2} g_-(\epsilon_2/v^{\sigma_2}) , \tag{94}$$

where v is given in (72). If $T < T_t$, (94) does not have any solution because $g_-(z)$ is finite and different from zero for $0 \leq -z \leq \infty$. This implies that the tricritical point is approached from the two-phase region in Fig. 6. If $T > T_t$, it is more convenient to use the representation (75) for M. Taking into account (77) it is found that $\Delta H = H - H_t$ is given by

$$\Delta H \sim \frac{T_t}{T'_{mt}} \epsilon_2 (1 + L' |\epsilon_2|^{(3-\tau_2)/\sigma_2}) , \quad (95)$$

where L' is a postive constant. This shows that the tricritical point is approached along a direction asymptotically parallel to the $T_m(H)$ line. The rate of approach to this direction is $|\epsilon_2|^{1+(3-\tau_2)/\sigma_2}$, and this is fast enough so that one obtains the same results as if the tricritical point were approached along the $T_m(H)$ line. In fact, the singular contributions to the different physical quantities are proportional to integrals of the form

$$\int_0^\infty d l_2 \, l_2^s \exp(-|\epsilon_2| l_2^{\sigma_2} - |\epsilon_2|^{1+(3-\tau_2)/\sigma_2} l_2)$$

$$\sim |\epsilon_2|^{-(1+s)\min[\sigma_2^{-1}, 1+(3-\tau_2)/\sigma_2]} = |\epsilon_2|^{-(1+s)/\sigma_2} , \quad (96)$$

where use has been made of the condition $(\tau_2 - 2)/\sigma_2 \leq 1$. We conclude that the renormalized exponents for M, χ, and C_{H_0} coincide with those given in (79) and (86) which in this case read

$$\zeta_X = (\tau_2 - 2)/\sigma_2 , \qquad \theta_X = (3 - \tau_2)/\sigma_2 ,$$

$$\bar{\alpha}_X = (1 + 2\sigma_2 - \tau_2)/\sigma_2 , \quad (97)$$

respectively. If σ_2 and τ_2 take values which are typical for σ and τ, $\bar{\alpha}_X$ is positive and of the order of α, and this contrasts with the finite cusp at $T_N(H)$ as given by (82). In case σ_2 and τ_2 satisfy relation (81), the renormalized specific heat C_{H_0} has an exponent $\bar{\alpha}_X = 1 - \mu^{-1}$ which is negative. Then the renormalized C_{H_0} remains finite at the tricritical point, but it has anomalies in its first- or higher-order derivatives depending on the value of μ. This corresponds to the behavior discussed by Griffiths[8] of the specific heat C_{px} of a He^3-He^4 mixture at constant pressure and composition near the tricritical point.

VII. SUMMARY AND CONCLUSIONS

We have shown that the ferromagnetic fluctuations induced in an antiferromagnet by an external magnetic field have a tendency at low temperatures to consist of compact ferromagnetic clusters if there is a ferromagnetic coupling between the spins of different sublattices. This is similar to what happens to the fluctuations of an antiferromagnet in zero external field, the only difference being that in this case the clusters are antiferromagnetic. In this situation the formula we propose with the inclusion both of F and AF clusters seems very sound, like the original droplet formula,[9] at low temperatures. This includes the low-temperature part of the metamagnetic transition which, therefore, can be identified with the locus of stability of ferromagnetic clusters. On the other hand, we are not so much interested in this region as in the high-temperature region around the tricritical point. The extrapolation of our droplet model to high temperatures is questionable, exactly as in the original model. The fact that the droplet model is satisfactory[7] in the case of ordinary critical points (in the sense that it gives a picture consistent with scaling) gives some confidence that the extrapolation has some validity in the present case also. The only reason why the extrapolation of the model at high temperatures should work less well when an external field is present might be the presence of an extremely strong interaction between the clusters of the two different kinds near the tricritical point. We have also found it necessary to take explicitly into account the effects of the interaction between clusters through population factors. Again this problem is strictly connected with the problem of a similar factor being present in the droplet formula for ordinary critical points.[11] In that case we have not been able to give a microscopic justification for this factor, but the effect which this factor tries to take into account is certainly present and the modified droplet formula gives a much better description of the fluctuations above T_c than the original model. Moreover, it is found[12] that the modified droplet formula is also rather satisfactory for both the amplitudes of the different singular quantities and the complete singular part of the equation of state in the neighborhood of the critical point for the Ising model in two and three dimensions. All this gives confidence in our droplet model even in the present case. Besides, we notice that our results do not depend on the particular form [Eqs. (17) and (18)] of the population factors as long as these factors depend on the same basic variables and have the property of becoming unity or zero, when $l \to \infty$, as for expressions (17) and (18). Our discussion has concerned the spin-$\frac{1}{2}$ Ising model, but it holds for strongly anisotropic Heisenberg models too. We expect, indeed, that our model describes, at least qualitatively, the tricritical point of other systems whenever the order is destroyed by fluctuations of two different kinds which consist of clusters of compact shape. In particular, this is the case for the spin-1 Ising model recently considered by Blume *et al.*[19] as a model for He^3-He^4 mixtures.

One of the main results of our model is that it predicts the existence of two different kinds of tricritical points with rather different critical behavior. In the first case the first-order metamagnetic transition ends at a tricritical point because of the effect of the antiferromagnetic clusters which become critical. The second possibility is that at the tricritical point both kinds of clusters become critical. The fluctuations for the former kind of tricritical point are much less pronounced than for the second kind, as shown by the stronger divergence of the susceptibility and of the specific heat in the last case. Another remarkable difference

is present for the difference $(M_+ - M_-)$ of the magnetizations of the two coexisting phases along the metamagnetic transition. In the first case this difference has essentially a linear behavior near the tricritical point, whereas in the second case it goes to zero with a power behavior like the order parameter at an ordinary critical point. From the experimental point of view this is probably the most easily observable characterization of the two kinds of tricritical points. The critical behavior is completely determined by the two exponents σ_1 and τ_1, characteristic of the system in zero field, by two exponents σ_2 and τ_2 relating to the ferromagnetic clusters, and by the exponent g which determines the variation of the microscopic surface free energy of antiferromagnetic clusters along $T_m(H)$. As a summary of our results, the critical behavior is given by (36) and (38) at $T_N(H)$ for both kinds of metamagnetic substances. For a tricritical point of first kind the critical behavior is given by (38) and (40) if H is kept constant and by (46)–(52) if (H_t, T_t) is approached along $T_m(H)$. For a tricritical point of second kind the critical behavior is given by (69) and (70) if $H = H_t$ and by (62)–(67) along $T_m(H)$. In this last case, if the interference effects between F and AF clusters are small, (79) and (80) apply and relations between the critical indices similar to the scaling laws[7] can be deduced. In the general case such relations do not exist. The renormalized critical behavior is given by (82), (83), and (97).

In comparison with the results of "classical" theories, the present theory is in disagreement on different counts, apart from predicting "nonclassical" exponents at the AF transition. In the first place, the Landau theory predicts only one kind of tricritical point, similar to the one we call the first kind, and does not predict a tricritical point of the second kind. In the case of a tricritical point of the first kind the main differences are the two following. The antiferromagnetic line in Fig. 5 need not, as in the Landau theory, be the continuation at the tricritical point of the paramagnetic line M_+. Moreover, the susceptibility is divergent upon approaching the tricritical point along M_+, whereas the Landau theory predicts no anomaly. In both cases our results are in better agreement with experiments and agree with the results implied by Griffiths's homogeneous form for the free energy.[8] However, the present picture of the tricritical point is rather different from that of Griffiths. In fact, for our model the free energy near the tricritical point of the first kind does not have a homogeneous form and the linear behavior of $M_\pm$ and the anomaly in χ come from different terms of the free energy. Griffiths's form for F is obtained in our model for a tricritical point of the second kind for a particular choice of the basic exponents τ_2 and σ_2 of the droplet formula if the effect of interference between F and AF clusters can be neglected. In fact, if $\sigma_2 = \tau_2 - 2$ then M_+ and M_- have a linear behavior as in Fig. 5. However, this relation among σ_2 and τ_2 seems rather unlikely to be present, in view of our physical picture of the system, even if it cannot be excluded in principle. The two possibilities we find for obtaining a linear behavior for $M_\pm$ can be distinguished on the basis of a behavior of χ and C_H. In fact, if the tricritical point is of the first kind the critical indices for χ and C_H must be the same as at the antiferromagnetic transition in finite magnetic fields, and only the relative amplitudes may be different. In the other case, the critical indices for χ and C_H are different.

We now compare the results of our model with the available experimental data. As already mentioned in the Introduction, the tricritical point in some magnetic systems, in particular in DAG,[3] seems to have a rather different character from that at the two-fluid critical mixing point of He^3-He^4, in spite of the fact that the two systems are thermodynamically analogous. $(M_+ - M_-)$ in DAG seems more likely to have a power behavior near T_t as in Fig. 6, whereas the analogous difference in composition $(x_+ - x_-)$ in He^3-He^4 has a linear behavior.[13] The specific heat C_{H_0} in constant external field of DAG has an anomaly at (H_t, T_t), whereas experiments[20] show no anomaly in C_{px} at the critical mixing point of He^3-He^4 mixtures. This suggests that the tricritical point in He^3-He^4 mixtures is of the first kind (or of the second kind with the atypical value $\sigma_2 = \tau_2 - 2$). More precise measurements of M in DAG, as well as in other metamagnets, are needed to confirm this difference. Also, precise measurements of the phase boundary near (H_t, T_t) and of the sublattice magnetization along $T_m(H)$ should be very useful. The absence of data close enough to the tricritical point of magnetic systems does no allow, unfortunately, a detailed comparison with the theory.

The most extensive data are those for DAG,[3] and they seem generically in agreement with our picture of a tricritical point of the second kind. As we said above, M_+ seems to have a power behavior as in Fig. 6 and, in any case, M_+ is not the continuation at (H_t, T_t) of the magnetization along $T_N(H)$. The specific heat C_{H_0} and the susceptibility has an anomaly at (H_t, T_t), and both the specific heat at constant internal field C_H and χ seem to have a stronger anomaly upon approaching the tricritical point than along $T_N(H)$. All these features are in qualitative agreement with our model. Keen *et al.*[3] interpret their data as showing evidence that the AF transition for $H \neq 0$ is of "higher-order" type because C_H and χ have finite peaks at $T_N(H)$. This is in disagreement with present theoretical views[21]

and, in particular, with the predictions of our model. However, in disagreement with that interpretation, the data in DAG seem consistent with our model. It is true that our model predicts a divergence for χ at $T_N(H)$, but this divergence is proportional to the specific heat. It is well known that for any λ point the anomaly of the specific heat is very mild so that the same should be expected for χ. The experimental data seem to be in agreement with our formula (34), at least semiquantitatively. As an example, from the experimental phase boundary[3] of DAG, we roughly estimate $\partial T_N(H)/\partial H \simeq -10^{-4}\,^\circ$K/Oe at $H = 0.75$ kOe and $\partial T_N(H)/\partial H \simeq -2.5\times 10^{-4}\,^\circ$K/Oe at $H = 2.0$ kOe. In zero external field the maximum value of C_H which has been observed[3] is ~ 5 R, so that from (34) we estimate that the maximum value we may expect for the singular part of χ is $\chi_{sing} \simeq 1.5\times 10^{-2}$ emu cm^{-3} at $H = 0.75$ kOe and $\chi_{sing} \simeq 8\times 10^{-2}$ emu cm^{-3} at $H = 2.0$ kOe. These values compare rather well with the experimental data[3] from which we estimate $\chi_{sing} = 0.03$ and 0.11 emu cm^{-3}, respectively. The shape of the anomaly of the specific heat C_H of DAG in finite fields has not yet been analyzed, but the data already show a field dependence of the amplitude of the anomaly along $T_N(H)$. This behavior can be interpreted on the basis of our model by a smooth temperature dependence of some of the constants appearing in the cluster formula, like B_1 in the population factor (17). In this case we should expect slight deviations from (37).

The tricritical point in He^3-He^4 mixtures can be interpreted as an example either of a tricritical point of the first kind or of the second kind but with the particular value $\sigma_2 = \tau_2 - 2$ and with small interference effects between clusters. The measurement of C_{px} is not able to distinguish between these two possibilities. In the first case C_{px} is expected to have a cusp (C_{px} is a "renormalized" quantity) and the disappearance[20] of the anomaly of C_{px} at the critical mixing point implies that the amplitude of the cusp vanishes at the tricritical point. In the second case the "renormalized" exponent is $(1-\mu^{-1})$, $0<\mu<1$, so that C_{px} might have, depending on the value of μ, a continuous first derivative. This corresponds, in general, to a flat maximum for C_{px}, and the experimental data require that the relative amplitude vanishes at the tricritical point. In both cases the vanishing of these amplitudes is not understood. Detailed measurements of the quantity corresponding to χ in the magnetic case, i.e., $\partial x/\partial(\mu_3 - \mu_4)$, where μ_3 and μ_4 are the chemical potentials of He^3 and He^4, might help in distinguishing between the two possibilities.

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