Tetracritical points in antiferromagnetic systems

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It is pointed out that certain XY-like antiferromagnets, including Cr_2TeO_6 and $KCuF_3$, should exhibit a *tetracritical* point at ($T = T_N$, $\vec{H} = 0$) when the magnetic field $\vec{H}$ is applied along a symmetry axis. The zero-temperature critical field is estimated to be of order 1 kG. The predictions of scaling and renormalization-group theories for tetracritical points may be tested experimentally on these antiferromagnets.

The phase diagram and critical behavior of systems which exhibit multicritical points (bicritical or tetracritical) have been of great interest in recent years.[1-5] Scaling theory and renormalization-group techniques have been applied to study the thermodynamic properties of a system near its multicritical point. These theories are found to be in excellent agreement with recent measurements on the uniaxial antiferromagnet $GdAlO_3$ near its bicritical point.[6] In general one expects that weakly anisotropic uniaxial antiferromagnets in a magnetic field $\vec{H}$, parallel to the easy axis, should exhibit a multicritical point at some finite nonzero value of H. Whether this point is *bicritical* or *tetracritical* is determined by the fourth-order anisotropy energy of the crystal.[2,4] Experimentally it is found that the anisotropy of uniaxial antiferromagnets is such that they tend to exhibit bicritical points in the (T,H) plane.

In the present note we discuss the (T,H) phase diagram of certain XY-like antiferromagnets (see Table I). We show that these compounds are expected to exhibit a multicritical point at $\vec{H}=0$, whose nature (namely, bicritical or tetracritical) is determined by the *direction* of the magnetic field. In particular we show that for $\vec{H}$ parallel to a certain symmetry direction the multicritical point becomes *tetracritical*, while for all other directions it is bicritical. We suggest that the various predictions of scaling and renormalization group-theories for the *tetracritical* point be tested experimentally on these antiferromagnetic systems. One expects that the experimental data near a tetracritical point occuring in zero field should be easier to interpret than in finite field because (a) the tetracritical point can be determined more accurately, since only the temperature need be varied, and (b) demagnetization corrections are not important since the interesting region of the phase diagram in the (T,H) plane lies close to the $H=0$ axis.

Consider, for example, a tetragonal crystal whose space group is $G_0 = P4/mmm$. We assume that it undergoes a phase transition which results in an antiferromagnetic structure, where ferromagnetic (001) planar layers are coupled antiferromagnetically. The sublattice magnetization is assumed to lie *in* the plane ferromagnetic

TABLE I. Antiferromagnetic compounds which are expected to exhibit $\vec{H}=0$ tetracritical point.

No.	Compound	T_N (°K)	Paramagnetic space group	Magnetic structure	Reference
1	Cr_2TeO_6	105	$P4_2/mnm$	Four magnetic ions per unit cell Type ^{12}G	12
2	Cr_2WO_6	69	$P4_2/mnm$	Four magnetic ions per unit cell Type ^{12}A	12,13
3	MnNi, MnPd MnPt	1073, 813 970	$P4/mmm$	Ferromagnetic (001) planes coupled antiferromagnetically	14
4	$KCuF_3$ $KCrF_3$	≤ 38	$I4/mcm$ or $P4mbm$	Ferromagnetic (001) planes coupled antiferromagnetically	15,16
5	Mn_2As	573	$P4/nmm$	Ferromagnetic (001) planes coupled antiferromagnetically	17
6	Fe_2As	323	$P4/nmm$	Ferromagnetic (001) planes ordered (+++−−−)	18

layers. Such a transition is thus described by an $(n=2)$-component vector model where the two components, A_x and A_y, of the order parameter correspond to the staggered magnetization in the x and y directions, respectively. The Landau-Ginzburg-Wilson (LGW) Hamiltonian which describes such a system is

$$-\frac{1}{kT}\mathcal{H}=-\int d^dx\{\tfrac{1}{2}r(A_x^2+A_y^2)+\tfrac{1}{2}[(\nabla A_x)^2+(\nabla A_y)^2] +u(A_x^4+A_y^4)+vA_x^2A_y^2\}, \quad (1)$$

where u and v are the only fourth-order terms invariant under the group G_0. The Hamiltonian might include nonisotropic gradient terms such as

$$\left(\frac{\partial A_x}{\partial x}\right)^2+\left(\frac{\partial A_y}{\partial y}\right)^2 \text{ or } \frac{\partial A_x}{\partial x}\,\frac{\partial A_y}{\partial y}.$$

It has been shown[7] that these terms are irrelevant; hence we do not include them in the Hamiltonian (1). Let us apply a small magnetic field $\vec{H}=(H_x,H_y,0)$ in the (x,y) plane. We assume, for simplicity, that $H_z=0$. However, since H_z does not break the symmetry of the Hamiltonian, our results remain valid when the applied field has a finite nonzero component in the z direction. The LGW Hamiltonian takes the form

$$-\frac{1}{kT}\mathcal{H}=\int d^dx\{\tfrac{1}{2}(r+aH_x^2+bH_y^2)A_x^2 +\tfrac{1}{2}(r+aH_y^2+bH_x^2)A_y^2 +\tfrac{1}{2}[(\nabla A_x)^2+(\nabla A_y)^2]+cH_xH_yA_xA_y +u(A_x^4+A_y^4)+vA_x^2A_y^2\}, \quad (2)$$

where a, b, and c are coupling terms allowed by the symmetry. We assume that the fourth-order terms do not significantly change with the field. This assumption simplifies the discussion but will not affect the results, as long as the applied field is small. The magnetic field acts as a symmetry-breaking parameter in the Hamiltonian (2), and the $\vec{H}=0$ critical point becomes a multicritical point in the $(T,\vec{H})$ space. Let us discuss the phase diagram of the Hamiltonian (2). Consider, first, the case where the magnetic field is applied along the x direction. In this case the coupling term $cH_xH_yA_xA_y$ vanishes, and the $H=0$ critical point becomes[4] bicritical or tetracritical depending on whether $v-2u$ is positive or negative, respectively (see Fig. 1). However we will show that if the magnetic field is applied along the [110] direction, one obtains a tetracritical point for $v-2u>0$ and bicritical point for $v-2u<0$. Let us define two variables:

$$A_+=(A_x+A_y)/\sqrt{2}, \quad A_-=(A_x-A_y)/\sqrt{2}. \quad (3)$$

For $\vec{H}=(1,1,0)H/\sqrt{2}$, the Hamiltonian (2) then takes the form

$$-\frac{1}{kT}\mathcal{H}=-\int d^dx\{\tfrac{1}{2}[r+\tfrac{1}{2}(a+b+c)H^2]A_+^2 +\tfrac{1}{2}[r+\tfrac{1}{2}(a+b-c)H^2]A_-^2 +\tfrac{1}{2}[(\nabla A_+)^2+(\nabla A_-)^2] +\tilde{u}(A_+^4+A_-^4)+\tilde{v}A_+^2A_-^2\}, \quad (4)$$

where $\tilde{u}=\frac{1}{4}(2u+v)$ and $\tilde{v}=\frac{1}{2}(6u-v)$. Note there is again no coupling term proportional to A_+A_-. Since $\tilde{v}-2\tilde{u}=-(v-2u)$, the phase diagram of the Hamiltonian (4) exhibits a tetracritical point whenever the phase diagram of the Hamiltonian (2), with $H_y=0$, exhibits a bicritical point, and vice versa. Therefore, any system which is described by the Hamiltonian (2) will exhibit a tetracritical point either for $\vec{H}\parallel[100]$ or for $\vec{H}\parallel[110]$. Tetracriticality is expected only if the magnetic field is applied *precisely* along one of the symmetry directions ([100] or [110]). When the field is applied in a general direction, the Néel point ($T=T_N$, $\vec{H}=0$) becomes a bicritical point in the (T,H) plane irrespective of the sign of $v-2u$. The phase diagram in the three-dimensional space (T,H_x,H_y) is sketched in Figs. 2(a)

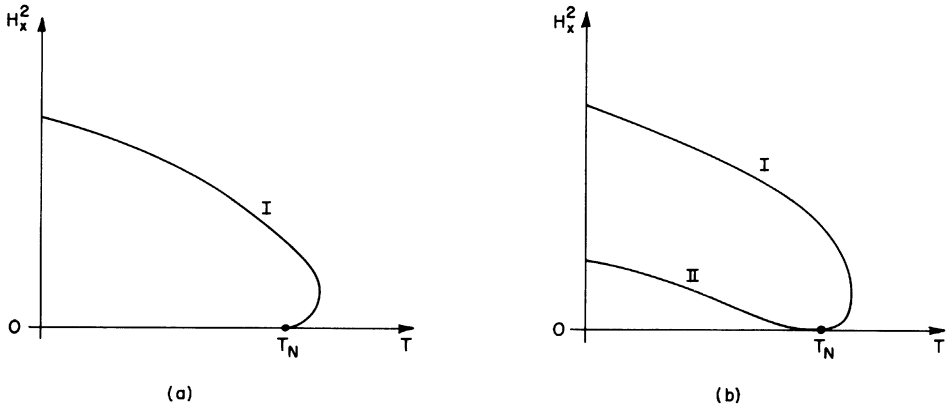

FIG. 1. (T,H_x) phase diagram corresponding to the Hamiltonian (2) for (a) $v-2u>0$, and (b) $v-2u<0$.

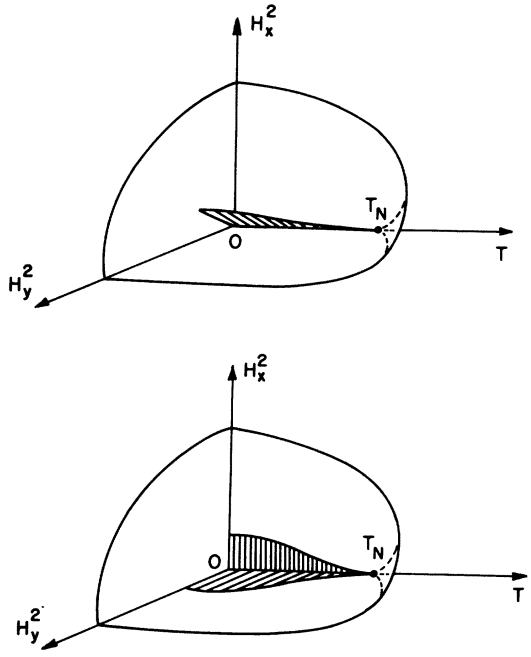

FIG. 2. (T, H_x, H_y) phase diagram corresponding to the Hamiltonian (2) for (a) $v-2u>0$, and (b) $v-2u<0$.

and 2(b) for $v-2u>0$ and $v-2u<0$, respectively. These figures display features similar to those found by Rohrer and Thomas,[8] and Fisher[9] for the phase diagram of uniaxial anisotropic antiferromagnets like $GdAlO_3$ in a field not parallel to the easy axis.

There exist many tetragonal crystals which, on the basis of the above discussion, may be expected to exhibit a tetracritical point at $\vec{H}=0$; some of these are listed in Table I. These compounds exhibit different antiferromagnetic structures but they may all be described by the same LGW Hamiltonian (2). One should, however, note that not every $n=2$ tetragonal crystal is expected to exhibit a tetracritical point at $\vec{H}=0$: in particular it is essential that the order parameter is not coupled linearly to the magnetic field, and that there exists a term $cH_xH_yA_xA_y$ in the Hamiltonian. An example of an $n=2$ tetragonal crystal which does not satisfy these requirements[10] is K_2NiF_4, whose LGW Hamiltonian does not, for reasons of symmetry, include a term $cH_xH_yA_xA_y$. The nature of the $\vec{H}=0$ multicritical point for this crystal is, therefore, not affected by the direction of the magnetic field; i.e., we predict that the phase diagram exhibits a bicritical point for $v-2u>0$ and a tetracritical point for $v-2u<0$ whatever the direction of $\vec{H}$.

The shape of the critical lines I and II near the tetracritical point [see Fig. 1(b)] has been studied

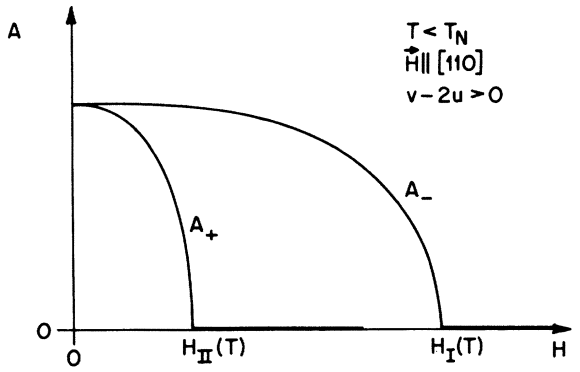

FIG. 3. Expected behavior of the order parameters A_+ and A_- as a function of the magnetic field parallel to the [110] axis.

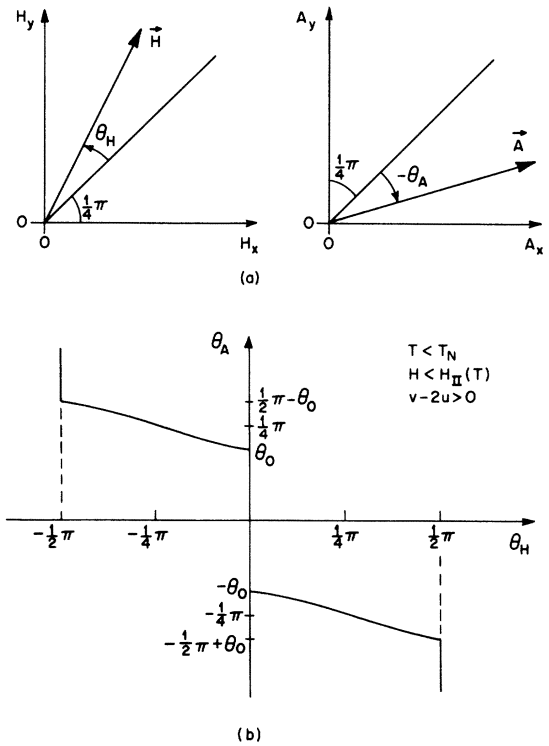

FIG. 4. Behavior of the orientation of the order parameter as a function of the orientation of the field when $v-2u>0$. (a) Definition of the orientation angles θ_H and θ_A; (b) relation between θ_H and θ_A for $H<H_{II}(T)$. Note that θ_A undergoes a spontaneous jump from θ_0 to $-\theta_0$ as θ_H passes through zero. In practice, of course, one might observe hysteresis. Corresponding jumps take place at $\theta_H=\pm\frac{1}{2}\pi$ and $\pm\pi$. The signs of θ_A and θ_H depend on the phenomenological parameters a, b, c. In the figure we assume that θ_A and θ_H have opposite signs since for most physical systems the antiferromagnetic component tend to align perpendicular to H, if the magnetic field is along the easy axis ($\theta_A=-\frac{1}{4}\pi$ for $\theta_H=\frac{1}{4}\pi$).

in detail by Bruce and Aharony.[4] They found that

$$t_{\mathrm{I}} \equiv [T_{\mathrm{I}}(H^2) - T_N]/T_N \sim (H^2)^{1/\psi_1} \tag{5}$$

and

$$t_{\mathrm{II}} \equiv [T_{\mathrm{II}}(H^2) - T_N]/T_N \sim (H^2)^{1/\psi_2}, \tag{6}$$

where $\psi_1 = \phi_{H^2}$ is the crossover exponent associated with the magnetic field[9] and $\psi_2 = \phi_{H^2} - \phi_v$, where ϕ_v is the crossover exponent associated with the cubic anisotropy. The ϵ-expansion expressions for the crossover exponents[2-4] indicate that $\psi_1 \simeq 1.16$ for $n = 2$ and $d = 3$, while $\psi_2 - \psi_1 \simeq 0.1$.

Let us now estimate the critical field H_{II} at zero temperature since this will determine the observability of the tetracritical lines II. The mean-field expressions for the critical fields $H_{\mathrm{I}}(T)$ and $H_{\mathrm{II}}(T)$ obtained from the Hamiltonian (4) are

$$H_{\mathrm{I}}^2(T) = 2\tilde{r}(T, H_{\mathrm{I}})/c\,, \tag{7}$$

$$H_{\mathrm{II}}^2(T) = -[2\tilde{r}(T, H_{\mathrm{II}})/c]\,[(2\tilde{u} - \tilde{v})/(2\tilde{u} + \tilde{v})]\,, \tag{8}$$

where $\tilde{r}(T,H) = r + \frac{1}{2}(a+b)H^2$. If we assume that a, b, and c are of the same magnitude we obtain

$$H_{\mathrm{II}}^2 \simeq \delta_v H_{\mathrm{I}}^2, \quad \delta_v = (2\tilde{u} - \tilde{v})/(2\tilde{u} + \tilde{v})\,. \tag{9}$$

If the cubic anisotropy[11] is of magnitude $\delta_v \simeq 10^{-1}$–10^{-2}, and the critical field H_{I} is of order 10 kG, the critical field H_{II} is found to be of magnitude about 1 kG. The phase diagram (see Fig. 1) can be studied experimentally by measuring, for example, the specific heat C_H and the direct susceptibility χ. As a function of T these are both expected to diverge like $t^{-\alpha}$ as the critical lines I and II are crossed. Similarly as a function of the field χ should display $(H - H_{\mathrm{I}})^{-\alpha}$ and $(H - H_{\mathrm{II}})^{-\alpha}$ peaks. Here $\alpha \simeq 0.12$ is the specific-heat exponent of the Ising model. Under bicritical conditions no singularities should be seen in the low-field region below T_N. The phase diagram can also be studied by direct measurement of the order parameters A_x and A_y using neutron-diffraction techniques. The expected behavior of some of the experimentally observable quantities as a function of the field is sketched in Figs. 3 and 4.

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[1] K. S. Liu and M. E. Fisher, J. Low Temp. Phys. 10, 655 (1973).
[2] M. E. Fisher and D. R. Nelson, Phys. Rev. Lett. 32, 1350 (1974).
[3] D. R. Nelson, J. M. Kosterlitz, and M. E. Fisher, Phys. Rev. Lett. 32, 1350 (1974); J. M. Kosterlitz, D. R. Nelson, and M. E. Fisher, Phys. Rev. B 13, 412 (1976).
[4] A. Aharony and A. D. Bruce, Phys. Rev. Lett. 33, 427 (1974); A. D. Bruce and A. Aharony, Phys. Rev. B 11, 478 (1975).
[5] D. R. Nelson and E. Domany, Phys. Rev. B 13, 236 (1976); J. M. Kosterlitz, J. Phys. C 9, 497 (1976); H. Horner, Z. Phys. B 23, 183 (1976).
[6] M. E. Fisher, Phys. Rev. Lett. 34, 1634 (1975); H. Rohrer, Phys. Rev. Lett. 34, 1638 (1975).
[7] A. Aharony and M. E. Fisher, Phys. Rev. B 8, 3349 (1973).
[8] H. Rohrer and H. Thomas, J. Appl. Phys. 40, 1025 (1969).
[9] M. E. Fisher, AIP Conf. Proc. 24, 273 (1975).
[10] R. J. Birgeneau, H. J. Guggenheim, and G. Shirane, Phys. Rev. B 1, 2211 (1970).
[11] A. H. Morrish, *The Physical Principles of Magnetism* (Wiley, New York, 1965), p. 317.
[12] M. C. Montmory and R. Newnham, Solid State Commun. 6, 323 (1968); W. Kunnman, S. La Placa, L. Corliss, and J. Hastings, J. Phys. Chem. Solids 29, 1359 (1968).
[13] M. C. Montmory, E. F. Bertaut, and P. Mollard, Solid State Commun. 4, 249 (1966).
[14] J. S. Kasper and J. Kouvel, J. Phys. Chem. Solids 11, 231 (1959); E. Krén and J. Solyom, Phys. Lett. 22, 273 (1966); L. Pál, E. Krén, G. Kadar, P. Sazbo, and T. Tarnoczi, J. Appl. Phys. 39, 538 (1968).
[15] M. T. Hutchings, E. J. Samuelsen, and G. Shirane, Phys. Rev. 188, 919 (1969).
[16] V. Scatturin, L. Corliss, N. Elliot, and J. Hastings, Acta Crystallogr. 14, 19 (1961). The paramagnetic space group of $KCrF_3$ is not well established.
[17] A. E. Austin, E. Adelson, and W. H. Cloud, J. Appl. Phys. Suppl. 33, 1356 (1962).
[18] H. Katsuraki and K. Suzuki, J. Appl. Phys. 36, 1094 (1965); H. Katsuraki and N. Achiwa, J. Phys. Soc. Jpn. 21, 2238 (1966).