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# Spin Flop, Supersolids, and Bicritical and Tetracritical Points

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A scaling theory is introduced for *bicritical* points, such as antiferromagnetic spin-flop points (with analogies to the upper $\lambda$ point in $^4$He), where two distinct critical lines meet. Experimentally testable predictions follow from renormalization-group calculations which indicate that the bicritical exponents should be Heisenberg like for systems with $n \leq 3$ components; the crossover exponent $\varphi$ ($\simeq 1.25$ for $n=3$) is directly observable. For $n>3$ an *intermediate* ("supersolid") low-temperature phase may appear, the bicritical point then becoming *tetracritical*.

The Hamiltonian of a uniaxially anisotropic antiferromagnet of $n$-component spins $\vec{S}(\vec{R}) = \{S_1(\vec{R}) \equiv S_\parallel(\vec{R}); \vec{S}_\perp(\vec{R})\}$ at the sites $\vec{R}$ of a $d$-dimensional lattice may be written

$$\mathcal{H}_{\rm int} = -\sum_{\vec{R},\vec{R}'} [J(\vec{R}-\vec{R}')\vec{S}(\vec{R})\cdot\vec{S}(\vec{R}') + D(\vec{R}-\vec{R}')S_\parallel(\vec{R})S_\parallel(\vec{R}')] - \sum_{\vec{R}} [H_\parallel S_\parallel(\vec{R}) + \vec{H}_\perp\cdot\vec{S}_\perp(\vec{R})] - \sum_{\vec{R}} \exp(i\vec{k}_0\cdot\vec{R})\vec{H}^\dagger\cdot\vec{S}(\vec{R}), \tag{1}$$

where $J(\vec{R})$ is the isotropic exchange coupling which leads to antiferromagnetic ordering on two interpenetrating but equivalent sublattices $A$ and $B$ (with superlattice reciprocal vector $\vec{k}_0$), while $D(\vec{R})$ (which might well vanish for $\vec{R}\neq 0$) represents the anisotropy energy aligning the spins along the "easy" or "parallel" axis. The staggered, ordering field $\vec{H}^\dagger = (H_\parallel^\dagger, \vec{H}_\perp^\dagger)$ is physically inaccessible, so that $\vec{H}^\dagger \equiv 0$, although its response may be studied by neutron diffraction. (In a real *bi*axial Heisenberg system, with one perpendicular "hard" axis, we may,[1] in the critical region, neglect the corresponding spin components and so take $n=2$ in place of $n=3$.)

In zero uniform external field ($\vec{H}=0$) the spins align parallel and antiparallel to the easy axis, and the critical point, at $T_{c0}$, should have Ising-like ($n=1$) character with corresponding Ising exponents visible for $T$ very close to $T_{c0}$.[1,2] If the uniform field $H_\parallel$ is now imposed (with $\vec{H}_\perp \equiv 0$, which in practice demands careful crystal alignment), one expects, as first pointed out by Néel,[3] that for $H_\parallel = H_\sigma(T) \sim DS$ the spin system should "flop" over, via a first-order transition, into an alignment that is predominantly perpendicular to the easy axis (see Fig. 1). The corresponding phase boundary in the $(H_\parallel, T)$ plane ends at what

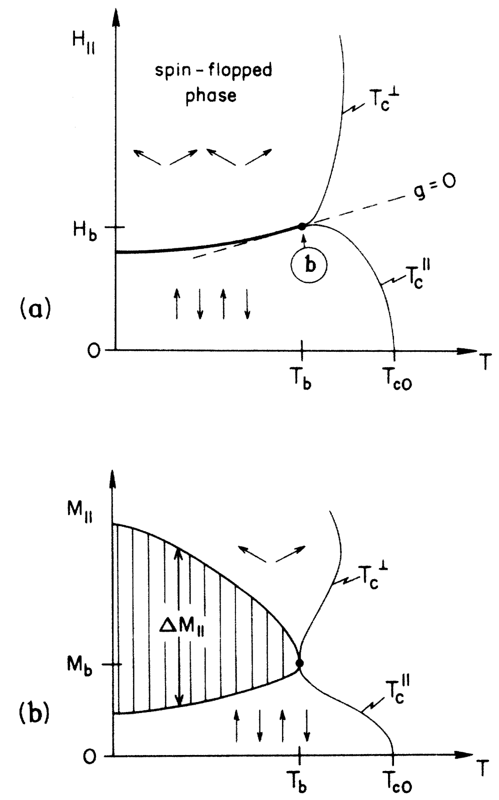


FIG. 1. (a) Schematic phase diagram of an anisotropic antiferromagnet in a uniform parallel field displaying a *bicritical* point. The bold line represents the locus of first-order spin-flop transitions. (b) Corresponding magnetization versus temperature diagram showing the jump $\Delta M_{\parallel}$ in magnetization (ruled lines).

may be called the "spin-flop point" ($H_b$, $T_b$) (see Fig. 1). For $H < H_b$ the line of critical points $T_c^{\parallel}(H_{\parallel})$ should be expected to remain of Ising ($n = 1$) character. However, for $H > H_b$ the relevant order parameter becomes $\vec{S}_{\perp}$ and hence the analogous line $T_c^{\perp}(H_{\parallel})$ should rather be expected to display distinct, $XY$-like critical behavior (for $n = 3$) or, more generally, $(n-1)$-isotropic character. Both these expectations (which we believe have not appeared explictly in the literature before) have been confirmed by explicit calculations[4] which will be summarized below.

Evidently the spin-flop point ($H_b$, $T_b$) lies at the join of two lines of critical points with distinct order parameters (and hence, in general, with distinct exponents, although, for example, both order parameters could be Ising like). Following the lead of Griffiths[5] we propose to name such points *bicritical* points.[6] At a bicritical point

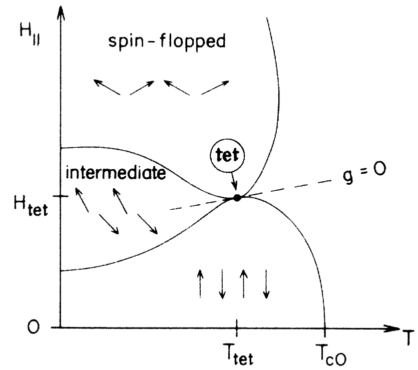


FIG. 2. Schematic phase diagram of an anisotropic ferromagnet exhibiting a *tetracritical* point and an intermediate phase with *both* parallel and perpendicular order.

two distinctly ordered phases both become identical with the totally disordered phase. Liu and Fisher[7] have presented a Landau-type phenomenological discussion of the competition between two order parameters which leads to such a bicritical point. However, they also discovered that for a certain range of phenomenological constants, a new "intermediate" phase could appear which *simultaneously* displayed *both* $\parallel$ and $\perp$ types of ordering. The bicritical point then became a *tetracritical* point[7] (see Fig. 2). In the context of pseudospin models for $^4$He (where $n = 3$ is appropriate) the spin-flop point corresponds to the upper $\lambda$ point.[7,8] An intermediate phase would then correspond to a "supersolid" phase in which both diagonal, crystalline, or "parallel" ordering and off-diagonal, superfluid, or "perpendicular" ordering simultaneously occur.[9] However, the renormalization-group analysis by Nelson, Kosterlitz, and Fisher[4] shows that such a tetracritical point with an intermediate phase can occur *only* if $n > n^{\times}(d)$, where $n^{\times}(3)$ probably exceeds 3 [although $n^{\times}(3) < 4$]. Since $n = 3$ for helium and for real antiferromagnets, this result provides theoretical justification for the apparent absence of a supersolid phase near the upper $\lambda$ point in helium, and possibly for the nonexistence, to our knowledge, of magnetic tetracritical points.

A *scaling theory* of a bicritical point (and, similarly, of a tetracritical point) may be formulated by introducing the reduced temperature and ordering fields,

$$t = \frac{T - T_b}{T_b}, \quad h_{\parallel} = \frac{H_{\parallel}^{\dagger}}{k_B T_b}, \quad h_{\perp} = \frac{H_{\perp}^{\dagger}}{k_B T_b}, \qquad (2)$$

and the modifying or deviating field,

$$g=(H_{\parallel}-H_b)/k_BT_b-j_bt,$$
$$j_b=[dH_\sigma/d(k_BT)]_b, \qquad (3)$$

which (see Fig. 1) measures the deviation of $H_{\parallel}$ from the tangent to the first-order phase boundary at the bicritical point. As usual the scaling postulate for the singular part of the free energy then takes the form

$$F_s(H_{\parallel},H_{\parallel}^{\dagger},H_{\perp}^{\dagger},T)\approx t^{2-\alpha}\mathfrak{F}\left(\frac{g}{t^{\phi}},\frac{h_{\parallel}}{t^{\Delta_{\parallel}}},\frac{h_{\perp}}{t^{\Delta_{\perp}}}\right). \qquad (4)$$

We must expect that the bicritical exponents $\alpha=\alpha_b$ and $\phi=\phi_b$ (and $\Delta_{\parallel}$ and $\Delta_{\perp}$) will differ from the corresponding exponents $\alpha_{\parallel}=\alpha_H(1)$, and $\alpha_{\perp}=\alpha_H(n-1)$, $\phi=\phi_H(n-1)$ on the critical lines. Here $\alpha_H(n)$ and $\phi_H(n)$ denote the usual specific heat and anisotropy crossover exponents of an isotropic, $n$-component Heisenberg-like model (with $n=1$, Ising, and $n=2$, $XY$); these are known numerically from series extrapolation (see, e.g., Refs. 2 and 8) and have been calculated in powers of $\epsilon=4-d$ by renormalization-group techniques.[1,10,11]

This scaling hypothesis may be justified by a renormalization-group argument.[10,12] Indeed, explicit calculations of the appropriate fixed points and exponents have been made[4] to order $\epsilon$. The analysis first confirms the expected $\parallel$ and $\perp$ nature of the exponents on the critical lines but, in addition, it shows that for $n\leq n^{\times}(d)\simeq(4+3.176\epsilon)/(1+1.294\epsilon)$ (up to order $\epsilon^3$ corrections), *the bicritical exponents are the same as those of a fully isotropic Heisenberg system,* i.e., $\alpha=\alpha_H(n)$, $\phi=\phi_H(n)$, $\Delta_{\parallel}=\Delta_{\perp}=\Delta_H(n)$. For $d=3$ the numerical evidence[2] thus yields the predictions $\alpha\simeq-0.10$ and $\phi\simeq1.25$ for $n=3$, and $\alpha\simeq0.02$ and $\phi\simeq1.18$ for $n=2$.

For $n>n^{\times}(d)$ a *new* fixed point of *biconical* symmetry[4] becomes stable under the renormalization group and all the exponents take on distinct, new values. Indeed, these biconical values, when expanded in powers of $\epsilon$, are found to have an *irrational* dependence on $n$ (in contrast to all previously examined cases where the coefficients are rational fractions in $n$).[4] More importantly, the corresponding fixed-point Hamiltonian satisfies the phenomenological conditions[7] for *tetracriticality* so that a doubly ordered, intermediate phase is then expected to appear (Fig. 2).

When $n$ crosses a higher borderline at $n=11+O(\epsilon)$ it is found[4] that the stable fixed point changes again from biconical to one which describes merely two *uncoupled* systems with *distinct* Ising-like and $(n-1)$-isotropic behavior. The scaling hypothesis (4) then no longer applies; instead the free energy is given asymptotically just by the sum of separate scaling forms describing the two distinct (and uncoupled) ordering processes. This distinct decoupling phenomenon has not previously been discovered in renormalization-group studies.

Various observable predictions follow directly from the scaling hypothesis (4): (A) The specific heat $C_{M_{\parallel}}$ on the locus $g=0$ (or on $M_{\parallel}=M_b$) diverges as $t^{-\alpha}$ (with cusp-like behavior[8] for $\alpha<0$); (B) the discontinuity in the magnetization across the spin-flop line below $T_b$ (see Fig. 1) varies as

$$\Delta M_{\parallel}(T)\sim t^{\tilde\beta} \text{ with } \tilde\beta=\beta_{M_{\parallel}}=2-\alpha-\phi, \qquad (5)$$

so that $\tilde\beta(n=3)\simeq0.85$ and $\tilde\beta(n=2)\simeq0.84$. For the first time this enables the anisotropy crossover exponent $\phi$ to be determined by direct observation of a power law.

(C) At $T=T_b$ the field deviation $|H_{\parallel}-H_b|\sim|g|$ varies as $|M_{\parallel}-M_b|^{\tilde\delta}$, with $\tilde\delta=\delta_{M_{\parallel}}=\phi/\tilde\beta$ which gives $\tilde\delta(n=3)\simeq1.47$ and $\tilde\delta(n=2)\simeq1.40$. (D) The direct susceptibility $\tilde\chi=(\partial M_{\parallel}/\partial H_{\parallel})_T$ for $g=0$ diverges as $t^{-\tilde\gamma}$ with $\tilde\gamma=\gamma_{M_{\parallel}}=2\phi+\alpha-2$ which, like (C), yields a cross check on scaling. Note that the values $\tilde\gamma(n=3)\simeq0.40$ and $\tilde\gamma(n=2)\simeq0.37$ contrast quite strongly with the weak singularities occurring in $\tilde\chi$ as the critical lines are approached *away* from the bicritical point; the behavior there matches[13] the corresponding specific heats $C_{H_{\parallel}}$, i.e., $\tilde\chi\sim t^{-\alpha_{\parallel}}$ or $t^{-\alpha_{\perp}}$ (although $C_{M_{\parallel}}$ and $C_{H_{\rm app}}$ are subject to standard exponent renormalization effects[14]). (E) The staggered or dominant ordering susceptibilities $\chi_{\parallel}$ and $\chi_{\perp}$ for $g=0$, observable in neutron scattering, diverge with exponents $\gamma_{\parallel}=2\Delta_{\parallel}+\alpha-2$ and $\gamma_{\perp}=2\Delta_{\perp}+\alpha-2$, which will again differ from those close to the critical lines (for $n=3$ we expect $\gamma_{\parallel}=\gamma_{\perp}\simeq1.38$; for $n=2$, $\gamma_{\parallel}=\gamma_{\perp}\simeq1.30$).

(F) The critical lines near the bicritical point are predicted to vary as

$$H_{\parallel}(T)\simeq H_b+(k_BT_bj_b)t\pm w_{\pm}t^{\phi}, \qquad (6)$$

where $w_+$ and $w_-$ are positive constants. If, as concluded above, $\phi>1$, this implies that the critical lines come in *tangent* to the first-order boundary. (G) Finally, in the $(M_{\parallel},T)$ plane the critical lines should vary as $t^{\tilde\beta}$ [see Fig. 1(b)].

Existing data[15] are not inconsistent with these various predictions but, unfortunately, they are insufficiently precise to confirm most of the details and, in particular, to determine exponents. However, the predicted tangency (F) of the critical lines at the bicritical point does seem to be indicated by the most detailed data of Shapira, Foner, and Misetich.[15] Further experiments which closely investigate the spin-flop point are clearly highly desirable and would provide an opportunity to check the crossover scaling predictions in an unusually direct fashion.

If the upper $\lambda$ point in $^4$He may be regarded as lying "close" to a bicritical point,[7,8] a possible explanation of the apparently nonuniversal specific-heat ratios, $A'/A$, observed by Ahlers on the $\lambda$ line at high pressure,[16] would lie in the crossover to bicritical behavior.

Although the biconical fixed point which describes tetracritical behavior does not seem to be relevant to real antiferromagnets, it may yet play a role in displacive transitions and at other types of polycritical points where the number of components of the residual order parameter can exceed $n = 3$.

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