proposed by Murao[14] may be formed at a temperature below 0.1 K. We plan to attempt to detect this ordering by neutron diffraction at lower temperatures and to investigate the mechanism of magnetic ordering in dilute PrNd alloys by inelastic scattering measurements on single crystals. The behavior of polycrystalline samples is not yet fully understood; presumably their magnetic ordering at relatively high temperatures is associated with strains and defects in the crystallites.

We have benefitted greatly from discussions with J. Jensen, P. A. Lindgård, B. Lebech, B. Rainford, P. E. Gregers-Hansen, M. Blume, and R. J. Elliott during the course of these experiments.

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Tetracritical Points in Mixed Magnetic Crystals*

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(Received 16 December 1974)

Recently developed renormalization-group techniques are used to study the tetracritical (or bicritical) point of quenched mixed magnetic crystals (e.g., A_pB_{1-p}, where A and B have ferromagnetic and antiferromagnetic phase transitions). The tetracritical (bicritical) exponents are possibly given by the $n=0$ value of their ϵ expansions. In particular, the crossover exponent φ, describing the shape of the p-T phase diagram near the tetracritical point, is *exactly* equal to 1. This explains various experiments.

Recent developments in the theory of critical phenomena led to a better understanding of phase diagrams which exhibit bicritical and tetracritical points.[1-4] Such points usually occur when a system has two order parameters, with competing interactions. A large class of such systems is that of quenched mixed magnetic crystals, of the type A_pB_{1-p}, when pure A and pure B have different types of magnetic ordering (e.g., ferromagnetic and antiferromagnetic). Examples are $Fe(Pd_pPt_{1-p})_3$, where $FePd_3$ is ferromagnetic and $FePt_3$ is antiferromagnetic,[5] $(Mn_{1-p}Fe_p)WO_4$, where pure $MnWO_4$ and $FeWO_4$ have distinct antiferromagnetic orderings,[6,7] $NH_4Cl_{1-p}Br_p$, where pure NH_4Cl and NH_4Br have parallel and antiparallel ordering of the ammonium tetrahedra,[8] UAs_pSe_{1-p}, where USe orders ferromagnetically while UAs orders antiferromagnetically,[9] and many others.

Unlike the anisotropic antiferromagnets in a uniform field, which tend to exhibit a *bicritical* spin-flop point and a first-order spin-flop line,[1] most of the mixed magnetic crystals exhibit a *tetracritical* point, and a mixed "intermediate"

ordered phase, bounded by two second-order lines (Fig. 1): For $p<p_t$ the transition at $T_1(p)$ is into an antiferromagnetic phase, and at $T_3(p)$ a ferromagnetic ordering is superimposed on the antiferromagnetic one. A similar picture occurs for $p>p_t$. Thus, it has been suggested[4] that these systems are ideal for studying recent theoretical predictions near tetracritical points. These predictions relate to the shape of the lines $T_1(p), \ldots, T_4(p)$ near p_t, to the nature of the transitions on each of these lines, etc.[1-4] The easiest one to check involves the relation $T_i(p)-T_t \sim g^{1/\psi_i}$, where (asymptotically close to the tetracritical point) $g \sim p-p_t$, and where the ψ_i's are shift exponents. Scaling and renormalization-group arguments yield[1-4] $\psi_1=\psi_2=\varphi$, where φ is the Riedel-Wegner crossover exponent for anisotropic systems.[10] Usually, $\varphi>1$, and therefore $T_1(p)$ and $T_2(p)$ approach T_t tangent to each other. The experiments on mixed crystals do not seem to yield this result.[5-8] Indeed, they seem to agree with the mean-field result, $\psi_1=\psi_2=1$.[7] The experimental situation as regards T_3 and T_4 is a little less clear.

In this note, I summarize preliminary results of a renormalization-group theory of mixed magnetic crystals, taking into account correctly both the randomness of the mixture[11] and the fact that both ferromagnetic and antiferromagnetic orders are possible.[12] The main result is that the tetracritical point may be described by exponents which are obtained from the ϵ expansion[13] by setting n, the number of spin components, equal to zero.[14] In this limit, the crossover exponent φ is *exactly* 1, as in mean-field theory, to all orders in $\epsilon=4-d$. Thus, the phase lines T_1 and T_2 in Fig. 1 are straight lines, in agreement with mean-field theory and with experiment, but at variance with all other predicted tetracritical and bicritical points.[1-4] However, other exponents are not mean-field-like. In particular, $\psi_3=\psi_4=1-\varphi_v>1$, where $\varphi_v=-\frac{1}{4}\epsilon+\frac{15}{64}\epsilon^2+O(\epsilon^3)$ is the "cubic" crossover exponent at $n=0$.[4] An experimental study of these may serve as a check of the theory, and yield much better understanding of tetracritical points.

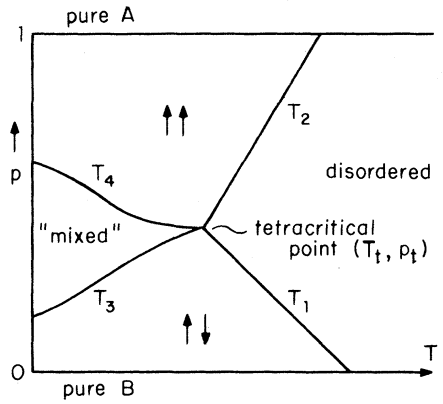

FIG. 1. Schematic phase diagram of the mixed magnetic crystal A_pB_{1-p}.

For simplicity, we consider here an Ising model, with the Hamiltonian

$$\mathcal{H}=-\sum_{\langle ij\rangle}\{J_{ij}^{AA}p_ip_j+J_{ij}^{AB}[p_i(1-p_j)+(1-p_i)p_j]+J_{ij}^{BB}(1-p_i)(1-p_j)\}S_iS_j, \tag{1}$$

where $p_i=1$ if the site i is occupied by an ion A, and $p_i=0$ otherwise. The sum is over site pairs $\langle ij\rangle$, and $J_{ij}^{\alpha\beta}$ denote the appropriate exchange integrals ($J_{ii}^{\alpha\beta}=0$). At this stage the argument is general; later we shall use a continuous-spin model, and the trace over the spin S_i will be understood as an integral over $-\infty<S_i<\infty$, with a weight function $\exp(-\frac{1}{2}S_i^2-vS_i^4-\ldots)$. For *quenched* crystals, the free energy is calculated as the average (over all possible spatial distributions $\{p_i\}$ of the two types of atoms) of the free-energy means.[11] The atoms are assumed to be immobile (on the relevant time scale) and the results to be independent of the particular distribution.

As a first step, we write $\mathcal{H}=\mathcal{H}_0+\mathcal{H}_1$, with

$$\mathcal{H}_0=-\sum_{\langle ij\rangle}\bar{J}_{ij}S_iS_j, \tag{2}$$

$$\mathcal{H}_1=-\sum_{\langle ij\rangle}[A_{ij}(p_i-p)(p_j-p)+D_{ij}(p_i+p_j-2p)]S_iS_j, \tag{3}$$

where $\bar{J}_{ij}$ is the coefficient of S_iS_j in (1) with all p_i's replaced by the average p, $A_{ij}=J_{ij}^{AA}-2J_{ij}^{AB}+J_{ij}^{BB}$, and $D_{ij}=J_{ij}^{AB}-J_{ij}^{BB}+A_{ij}p$. We now expand the free energy in a perturbation series in $\mathcal{H}_1$, and then average, term by term, over all configurations $\{p_i\}$ (assuming that the occupation probabilities of different sites are independent of each other[11]). The resulting free energy is

$$-\beta F=\ln \mathrm{tr}\exp(-\beta\mathcal{H}_0)+\sum_{\langle ij\rangle}\sum_{\langle il\rangle}\beta C_{ijl}[\langle S_iS_jS_iS_l\rangle-\langle S_iS_j\rangle\langle S_iS_l\rangle]+\ldots, \tag{4}$$

with $C_{ijl}=\beta p(1-p)[p(1-p)A_{ij}{}^2\delta_{jl}+2D_{ij}D_{il}]$, and the angular brackets denoting a thermal average with $\exp(-\beta\mathcal{H}_0)$. Following de Gennes,[14] and Grinstein and Luther,[11] we then note that the same free energy per degree of freedom will arise from the Hamiltonian

$$\mathcal{H}_{\rm eff}=-\sum_{\langle ij\rangle}\bar{J}_{ij}\sum_{\alpha}S_i{}^{\alpha}S_j{}^{\alpha}-\sum_{\langle ij\rangle}\sum_{\langle il\rangle}C_{ijl}\sum_{\alpha\beta}S_i{}^{\alpha}S_j{}^{\alpha}S_i{}^{\beta}S_l{}^{\beta}+\cdots, \tag{5}$$

where now $\{S_i{}^{\alpha},\ \alpha=1,\ldots,n\}$ is an n-component spin vector, if we set $n=0$ at the end of the calculation. This will eliminate terms like $\sum_{\alpha\beta}\langle S_i{}^{\alpha}S_j{}^{\alpha}\rangle\langle S_i{}^{\beta}S_l{}^{\beta}\rangle$ from the free energy, as required by (4). A similar analysis may be performed for higher-order terms in the spins, and to all orders in perturbation theory. Using the continuous-spin model near four dimensions, we need not consider the explicit form of these higher-order terms.[13]

We next want to discuss a possible simultaneous ferromagnetic and antiferromagnetic ordering. As recently shown by Nelson and Fisher,[12] this can be done by introducing two spin fields, $\sigma_+{}^{\alpha}(\vec{q})$ and $\sigma_-{}^{\alpha}(\vec{q})$, via

$$\sigma_{\pm}{}^{\alpha}(\vec{q})=\tfrac{1}{2}c_{\pm}[\sum_{i\subset\Gamma}\exp(i\vec{q}\cdot\vec{X}_i)S_i{}^{\alpha}\pm\sum_{i\subset\Omega}\exp(i\vec{q}\cdot\vec{X}_i)S_i{}^{\alpha}], \tag{6}$$

where $c_{\pm}$ are normalization constants, and Γ and Ω are the two interpenetrating sublattices into which the lattice is decomposed when the spins order antiferromagnetically. The wave vectors $\vec{q}$ run over a reduced Brillouin zone, corresponding to a superlattice. In terms of these variables, with appropriate choices of values for $c_{\pm}$, the partition function becomes

$$Z=\int\prod_{\vec{q}}[d^n\sigma_+(\vec{q})d^n\sigma_-(\vec{q})]\exp\mathcal{H},$$

with

$$\begin{aligned}\mathcal{H}=-\tfrac{1}{2}\int d^dx\{&(\nabla\vec{S}_+)^2+(\nabla\vec{S}_-)^2+r_+|\vec{S}_+|^2+r_-|\vec{S}_-|^2+2u_{++}|\vec{S}_+|^4\\&+4u_{+-}|\vec{S}_+|^2|\vec{S}_-|^2+2u_{--}|\vec{S}_-|^4+\textstyle\sum_{\alpha}[2v_{++}(S_+{}^{\alpha})^4+4v_{+-}(S_+{}^{\alpha})^2(S_-{}^{\alpha})^2+2v_{--}(S_-{}^{\alpha})^4]\},\end{aligned} \tag{7}$$

where we use schematic real-space notation, $\vec{S}_{\pm}(\vec{x})$ denoting the Fourier transform of $\vec{\sigma}_{\pm}(\vec{q})$. All the coefficients in (7) are simple functions of T and p. In particular, r_+ and r_- are combinations of $\bar{J}_{\Gamma}=\sum_{i\subset\Gamma}\bar{J}_{0i}$ and of $\bar{J}_{\Omega}=\sum_{i\subset\Omega}\bar{J}_{0i}$, where $0\subset\Gamma$. If $\bar{J}_{\Gamma}>0$ and $\bar{J}_{\Omega}<0$, then the equation $r_+=r_-$ has a solution $0<p_t<1$ for some temperature T_t.

For $p<p_t$, r_+ is greater than r_- and σ_- orders first to yield an antiferromagnetic ordering. This phase transition will be of the type discussed by Grinstein and Luther[11]: At $p=0$, the transition is Ising-like. For $p\gtrsim 0$, the impurities lead to a term $u_{--}\sum_{\alpha\beta}(S_-{}^{\alpha})^2(S_-{}^{\beta})^2$, and this finally leads to a first-order or smeared transition ($u_{--}<0$) or to a fixed point characterized by the $n=0$ exponents ($u_{--}>0$). Which of these will occur at $d=3$ depends on details of irrelevant parameters, which affect the first few iterations.[15] For p near 1, $r_+<r_-$ and a ferromagnetic ordering results with a similar behavior. For $p=p_t$, these two second-order lines meet at a bicritical or tetracritical point.[1-4] This point will be described by the stable fixed point of the renormalization-group recursion relations for the parameters in (7), when $r_+\simeq r_-$. These recursion relations yield many fixed points and not all of these have yet been fully analyzed.[16] However, it seems that probably only one fixed point, namely $v_{++}=v_{+-}=v_{--}=0$, $u_{++}=u_{+-}=u_{--}=\pi^2\epsilon/4+O(\epsilon^2)$, is fully stable. This is simply the analytic continuation of the fixed point describing the isotropic $2n$-component spin model[17] to $n=0$. To derive the shift (crossover) exponent from n-component to m-component behavior, we follow Bruce's[18] analysis: A diagram contributes to his expression for $T_c(g_A)$ (g_A is the anisotropy) only if it has at least one internal line involving r_2 [the $(n-m)$-component nonzero inverse susceptibility] and two external lines involving r_1 ($=0$). Such internal lines must be on closed loops, and yield prefactors of $n-m$.[19] With subtraction of $T_c(0)$, each such integral becomes of order r_2. The self-energy equation for r_2 shows that $(n-m)r_2$ is of order ng_A. Hence, in the limit $n\to 0$, all diagrams give zero contribution. Thus, the crossover to the lines T_1 and T_2 is given by $T_i(p)-T_t\sim p-p_t$. All other exponents are similarly obtainable from the ϵ expansion. In particular, the crossover exponent φ_v due to the (irrelevant) parameter $u_1=\frac{1}{2}(u_{++}+u_{--})-u_{+-}$, which generates terms like $\vec{S}_+{}^4+\vec{S}_-{}^4$, is given by the $n=0$ value of the "cubic" crossover exponent.[4] As shown by Bruce and Aharo-

ny,[4] the sign of this parameter determines if the point (T_t, p_t) is bicritical ($u_1 < 0$) or tetracritical ($u_1 > 0$). The fact that u_1 is irrelevant leads to values of $\psi_3 = \psi_4 = \varphi - \varphi_v$ which are larger than unity, so that the mixed phase is to be observed only at finite distances below T_t.

It should be emphasized that the recursion relations we study in the vicinity of $d = 4$ cannot give accurate information as regards the actual Hamiltonian flow at $d = 3$. Higher-order spin terms, and other irrelevant variables, affect the initial iterations. It is thus difficult to predict if a given Hamiltonian will indeed flow to the $n = 0$ fixed point and what will be the effective sign of u_1. In some cases, the flow may lead to anomalous regions, associated with smeared or first-order transitions,[15] or to some other stable fixed points (presumably with $\varphi > 1$).[16] What I want to emphasize here is that the $n = 0$ fixed point is stable, and *may* describe the tetracritical (bicritical) behavior for some (unknown) range of the initial parameters. The agreement with experiments seems to encourage belief in this possibility.

There are many possible generalizations of the present model: One can consider an anisotropic antiferromagnet, with an impurity which has an opposite anisotropy, leading to a metamagnetic tricritical point or a spin-flop point at lower uniform magnetic fields[9]; one can consider impurities which introduce anisotropies; etc.[16]

The author is grateful to A. H. Luther, D. R. Nelson, and M. E. Fisher for many helpful discussions, and especially for information on their work prior to publication. He also thanks P. C. Martin and H. Ehrenreich for their hospitality at Harvard University.

*Work supported in part by the National Science Foundation under Grant No. GH-32774.

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