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Decoupled Tetracritical Points in Quenched Random Alloys with Competing Anisotropies*

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The phase diagram of a quenched random alloy of two components with competing anisotropies exhibits two critical lines, corresponding to ordering of only m_1 (or m_2) spin components. The two lines meet at a tetracritical point, where all $m = m_1 + m_2$ components order simultaneously. General scaling arguments are used to show that the critical behavior at this point is *decoupled*, i.e., the m_1 - and m_2 -component subsystems have separate scaling free energies. Applications and experiments are briefly discussed.

In a recent Letter,¹ one of us studied the tetracritical point which arises in a random quenched mixture of ions with competing ferromagnetic and antiferromagnetic exchange interactions. Within the context of renormalization-group recursion relations near four dimensions, consequences were drawn from the assumption that a certain "isotropic n = 0" fixed point describes this tetracritical point. However, it was emphasized, that this assumption is probably never satisfied, because its parameters are "unphysical."² In this Comment we (a) generalize the physical problem of Ref. 1 to that of general randomly mixed alloys with competing spin anisotropies, (b) use general scaling arguments (not limited to the ϵ expansion) to study the stability of several simple fixed points, (c) conclude that a "decoupled" fixed point, at which the free energy is the sum of two separate scaling functions (for each of the competing order parameters) may describe the tetracritical point, and (d) report on some renormalization-group recursion relation studies which indicate that only the "decoupled" fixed point is stable in the "physical" region. Such "decoupled" fixed points were previously found only for pure systems with high spin dimensionality.^{3,4} We find that this behavior is the rule once randomness is introduced.

The Hamiltonian of anisotropic spin systems may be written

$$\mathcal{K} = -\sum_{\langle ij \rangle} \left\{ J_{ij} \vec{\mathbf{S}}(i) \cdot \vec{\mathbf{S}}(j) + D_{ij} \left[m_1^{-1} \vec{\mathbf{S}}_1(i) \cdot \vec{\mathbf{S}}_1(j) - m_2^{-1} \vec{\mathbf{S}}_2(i) \cdot \vec{\mathbf{S}}_2(j) \right] \right\},$$
(1)

where $\vec{S}(i) = \{\vec{S}_1(i); \vec{S}_2(i)\}\$ is an *m*-component spin vector at the site *i* of a *d*-dimensional lattice. \vec{S}_1 and \vec{S}_2 are m_1 - and m_2 -component parts of \vec{S} $(m_1 + m_2 = m)$. J_{ij} is an isotropic exchange coupling, while D_{ij} introduces an anisotropy aligning the spins in the \vec{S}_1 $(D_{ij} > 0)$ or the \vec{S}_2 $(D_{ij} < 0)$ subspaces. This Hamil-

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tonian has recently been used as the basic model for describing systems exhibiting bicritical and tetracritical points.^{1,3-6} As function of $D_{i,j}$ the system exhibits m_1 -component $(D_{ij} > 0), m_2$ -component $(D_{ii} < 0)$, or *m*-component $(D_{ii} = 0)$ Heisenberg-like critical behavior. The crossover from *m*-component to m_1 -(m_2)-component behavior is described by a scaling theory,^{1,7-9} which is supported by renormalization-group studies.^{8,9,4-6} In the present Comment, we wish to concentrate on the phase diagram which results from the variation of D_{ii} by randomly mixing materials with different values of D_{ij} .¹ The average \overline{D}_{ij} is thus a function of the relative concentration of the two mixed materials. If the two pure components have opposite signs of D_{ii} then we expect a phase diagram as exhibited in Fig. 1.^{1,10,11} In fact, such a phase diagram seems to be indicated by recent experiments on $K_2Mn_{a}Fe_{1-a}F_{4}$,¹² and by various experiments reviewed in Ref. 1. The shape of such phase diagrams has been the subject of much recent study.^{1,3-6,13} The usual result is that the critical lines meet tangentially at the multicritical point. However, it has been conjectured¹ that this may not be the case for random mixtures. Indeed, at the "decoupled" fixed point each subsystem orders at its own critical temperature, without being affected by the other subsystems. This leads to the two crossing lines in Fig. 1, which are roughly straight near the tetracritical point, where they meet at an angle. Some experimental information on mixed magnets seems to support this pic-



FIG. 1. Phase diagram of a random mixture of two components, tending to align the spins in the m_1 -component subspace $(D_{ij} > 0)$, with concentration p) or in the m_2 -component subspace.

ture.¹ An experimental study of the actual detailed new scaling behavior in this vicinity now becomes of great interest.

The argument is based on a trivial generalization of the by now standard n - 0 trick.¹⁴⁻²⁰ The free energy per spin component of the Hamiltonian (1) is the same as the n - 0 limit of the free energy per component of the *nm*-component Hamiltonian $\mathcal{H}_{eff} = \mathcal{H}_0 + \mathcal{H}_1$. Here, \mathcal{H}_0 is the same as \mathcal{H} in (1), except that $\mathbf{\bar{S}}$, $\mathbf{\bar{S}}_1$, and $\mathbf{\bar{S}}_2$ are replaced by $\overline{\sigma} = (\mathbf{\bar{S}}^1, \mathbf{\bar{S}}^2, \dots, \mathbf{\bar{S}}^n)$, $\overline{\sigma}_1 = (\mathbf{\bar{S}}^1, \mathbf{\bar{S}}_1^2, \dots, \mathbf{\bar{S}}_1^n)$, etc., while \mathbf{J}_{ij} and D_{ij} are replaced by their configurational averages \overline{J}_{ij} and \overline{D}_{ij} . If only the variable D_{ij} is random, and has a correlationless Gaussian distribution with second cumulant Δ_D for each bond ij, then

$$\mathcal{H}_{1} = \sum_{\langle ij \rangle} \left\{ u_{11} [\vec{\sigma}_{1}(i) \cdot \vec{\sigma}_{1}(j)]^{2} + 2u_{12} [\vec{\sigma}_{1}(i) \cdot \vec{\sigma}_{1}(j)] [\vec{\sigma}_{2}(i) \cdot \vec{\sigma}_{2}(j)] + u_{22} [\vec{\sigma}_{2}(i) \cdot \vec{\sigma}_{2}(j)]^{2} \right\},$$
(2)

with $u_{11} = -\Delta_D / 2k_B Tm_1^2$, $u_{12} = -m_1 u_{11} / m_2$, and $u_{22} = (m_1 / m_2)^2 u_{11}$. If J_{ij} is also random, then \mathcal{H}_1 remains unchanged in form, and only the coefficients u_{ij} assume new values. If the distribution is not Gaussian, then higher powers of the spins appear in (2). For our purposes it is sufficient to consider the leading terms, which already appear in (2). The higher-order terms are less relevant.²⁰

If $\mathcal{K}_1 \equiv 0$, then the Hamiltonian \mathcal{K}_0 reduces to *n* decoupled identical nonrandom Hamiltonians of the form (1). For $\overline{D}_{ij} = 0$, these lead to isotropic *m*-component critical behavior. If $\overline{D}_{ij} > 0$, these imply isotropic nonrandom m_1 -component behavior. If $\mathcal{K}_1 \neq 0$, then only the first term in \mathcal{K}_1 must now be considered. This is exactly the term considered previously for random isotropic ex-

change.¹⁶⁻¹⁸ A typical term here is $[\vec{S}_1^{\alpha}(i) \cdot \vec{S}_1^{\alpha}(j)] \times [\vec{S}_1^{\beta}(i) \cdot \vec{S}_1^{\beta}(j)]$ with $\alpha \neq \beta$. Near $u_{11} = 0$, each factor here scales as the energy density of an m_1 -component substem, i.e., as $\xi^{-(1-\alpha_{m_1})/\nu_{m_1}}$, where ξ is the correlation length (the unperturbed Hamiltonian now has *n* independent identical m_1 -component models). Thus, the crossover exponent related to u_{11} , φ_{11} , is found from the equation¹⁹

$$d - \varphi_{11} / \nu_{m_1} = 2(1 - \alpha_{m_1}) / \nu_{m_1}, \qquad (3)$$

which yields $\varphi_{11} \equiv \alpha_{m_1}$. Note that we do not expand the free energy in $\langle \mathcal{H}_1 \rangle$, or use any other perturbative procedure. Instead, we *exactly identify* the operator appearing in \mathcal{H}_1 as a *scaling eigenoperator* near the appropriate fixed point, with the eigenvalue $\lambda_{11} = \alpha_{m_1} / \nu_{m_1}$.¹⁹ The only input is the knowledge that the energy density is such an eigenoperator. One thus expects the random m_1 -component system to have the usual nonrandom behavior if $\alpha_{m_1} < 0$, i.e., for XY or Heisenberg systems at d=3. If $\alpha_{m_1} > 0$, as it is for $m_1=1$ at d=3, one finds a crossover to a new, "random" behavior. This new behavior has only been studied to order ϵ^2 , where $\epsilon = 4 - d$, if $m_1 > 1$,^{16,17} and to order $\epsilon^{1/2}$, if $m_1 = 1$.¹⁸ In both cases, the new exponent α is negative.

If $\overline{D}_{ij}=0$, then \mathcal{K}_0 corresponds to isotropic *m*component behavior. The interaction \mathcal{K}_1 , with the coefficients as given following (2), now has terms like the product $[m_1^{-1}\dot{\mathbf{S}}_1^{\alpha}(i)\cdot\dot{\mathbf{S}}_1^{\alpha}(j)-m_2^{-1}$ $\times\dot{\mathbf{S}}_1^{\alpha}(i)\cdot\dot{\mathbf{S}}_2^{\alpha}(j)][m_1^{-1}\dot{\mathbf{S}}_1^{\beta}(i)\cdot\dot{\mathbf{S}}_1^{\beta}(j)-m_2^{-1}\dot{\mathbf{S}}_1^{\beta}(i)$ $\cdot \mathbf{S}_2^{\rho}(j)]$. For $\alpha \neq \beta$, each of these factors scales as $\xi^{-(d^-\varphi_m/\nu_m)}$, where φ_m is the anisotropic exchange crossover exponent.⁷⁻⁹ Hence, the crossover exponent corresponding to Δ_D is ¹⁹ $(2\varphi_m - d\nu_m) = 2\varphi_m + \alpha_m - 2$, which is positive and of order 0.35 for d=3, m=2, 3.¹⁹ We thus must consider alternative fixed points.

A simple additional fixed point arises if $u_{12}^* = 0$. In this case, \mathcal{K}_{ff} separates into two independent parts, involving only the nm_1 -component vector $\vec{\sigma}_1$ or the *nm*₂-component vector $\vec{\sigma}_2$. The critical behavior of each of these will be characterized by the appropriate fixed point, which is the usual "pure," or "nonrandom" one if $\alpha_{m_i} < 0$ or the "random" one if $\alpha_{m_i}^{(pure)} > 0$. In the latter case, the "random" behavior also has a negative α (and a negative u_{ii}^{*}). We now consider the stability of this "decoupled" behavior with respect to the variable u_{12} . Again, u_{12} multiplies an operator which is the product of two independent energy operators, one corresponding to an m_1 -component system and the other corresponding to an m_2 -component system. Thus, it scales as $\xi^{-(1-\alpha_{m_1})/\nu_{m_1}-(1-\alpha_{m_1})/\nu_{m_1}}$, leading to a "crossover" exponent (φ/ν) equal to²¹ $(\alpha_{m_1}/\nu_{m_1} + \alpha_{m_2}/\nu_{m_2})/2$. Since both exponents α_{m1} and α_{m2} are *negative* at the stable fixed point, we conclude that the decoupled behavior is *stable* against the parameter *u*₁₂.

This kind of argument is easily generalized. Any system with two competing order parameters, with an energy-energy type of coupling, will have a decoupled tetracritical point if the specific heats of each order parameter separately does not diverge at T_c .

To complete the identification of the tetracritical point with the "decoupled" fixed point, we must study all other possible fixed points and all possible Hamiltonian flows under renormalizationgroup recursion relations. First, note that initially u_{11} and u_{22} are negative, while u_{12} is nonnegative. If J_{ij} were also random, then the signs of u_{11} and u_{22} would not change, but that of u_{12} may change. Thus, there certainly exist initial distributions with $u_{12} = 0$, which are described by the "decoupled" fixed point. Moreover, the flows for small u_{12} clearly go to this fixed point, as argued above. This shows the relevance of the results presented here to real systems.

More generally, one must now use the continuous spin model near four dimensions. This introduces three more variables, i.e., v_{11} , v_{12} , and v_{22} into \mathcal{H}_1 [the scalar products, like $(\vec{\sigma}_1 \cdot \vec{\sigma}_1)^2$, are replaced by $\sum_{\alpha} (\vec{s}_1 \cdot \vec{s}_1^{\alpha})^2]^{1}$ We have studied the recursion relations in the resulting six-dimensional parameter space. For v_{12} , $u_{12} > 0$, these yield no fixed point in the physical region $(u_{11}, u_{22} < 0)$. For $v_{12} = 0$, they yield 32 simple fixed points, of the types discussed above. Of these, only the "decoupled" one is stable. For $v_{12} \neq 0$, we thus far find numerically 22 ($m_1 = m_2$) = 1) or 23 $(m_1 = 1, m_2 = 2)$ additional fixed points. Of these, none of the physical ones is stable.²² We thus conclude that only the "decoupled" fixed point is appropriate to describe the tetracritical point.

The model discussed here, with only two possible directions of anisotropy, is distinct from that discussed in Ref. 19, in which the local anisotropy can continuously point along any direction in space. In that model, no stable ferromagnetic fixed point was found, and the transition probably becomes of a spin-glass nature.²³ A similar situation may sometimes occur when the two components of the mixture have competing ferromagnetic and antiferromagnetic ordering, which on the average exactly cancel each other, corresponding to $\overline{J}_{ij} = 0$ in the above discussion. In all other situations, the decoupled fixed point seems to describe the critical behavior near the tetracritical point, at least for a range of parameters in its vicinity. This may be the explanation for the agreement of experiments on rare-earth mixtures mentioned in Ref. 10, and those mentioned in Ref. 1, with Fig. 1.

One of us (A.A.) acknowledges a useful discussion with Dr. Per-Anker Lingård.

²The "isotropic n = 0" fixed point is irrelevant if it

^{*}Supported by a grant from the United States Israel Binational Science Foundation (BSF), Jerusalem, Israel.

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can never be reached under renormalization-group recursion relation interactions. This was shown to be the case in a simple situation [A. Aharony, Y. Imry, and S. Ma, Phys. Rev. B <u>13</u>, 466 (1976)], but has not been proved generally. Physical arguments [T. C. Lubensky, Phys. Rev. B <u>11</u>, 3573 (1975)] strongly indicate that this is generally true.

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These are very special cases, and we shall not consider them here. They do not apply to the Gaussian case, when (2) is exact.

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²²A detailed description of these results, and various extensions, will be reported elsewhere.

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ERRATUM

INELASTIC COLLISION INDUCED BY INTENSE OPTICAL RADIATION. D. B. Lidow, R. W. Falcone, J. F. Young, and S. E. Harris [Phys. Rev. Lett. 36, 462 (1976)].

Further investigations have indicated that the results reported in this Letter do not demonstrate a laser-induced inelastic collision. The experiment was not able to distinguish between a Sr $4d^{3}D - 5p^{3}F$ transition at 6408.5 Å and the Sr-Ca transfer predicted at 6408.6 Å.

We have subsequently performed two new exper-

iments in Sr-Ca which do not have such a wavelength coincidence [See S. E. Harris, R. W. Falcone, W. R. Green, D. B. Lidow, J. C. White, and J. F. Young, in *Tunable Lasers and Applications*, edited by A. Mooradian, T. Jaeger, and P. Stokseth (Springer, New York, 1976), p. 193, and R. W. Falcone, W. R. Green, J. C. White, J. F. Young, and S. E. Harris, "Observation of Laser Induced Inelastic Collisions" (to be published).] In both cases a laser-induced collision was observed, and the transfer cross section maximized at the expected interatomic wavelength.