Spin-Glass and Ferromagnetic Behavior Induced by Random Uniaxial Anisotropy

Robert A. Pelcovits,[a] E. Pytte, and Joseph Rudnick[b]
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598
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The effect of a random uniaxial anisotropy axis on the magnetic properties of an amorphous ferromagnet is considered. It is found that in a quenched system random anisotropy with an isotropic angular distribution destroys the ferromagnetic state in fewer than four dimensions. The low-temperature phase is, instead, an Edwards-Anderson spin-glass. In more than four dimensions ferromagnetism or spin-glass ordering is found depending on the degree of disorder. Some properties of the ferromagnetic state are outlined.

In this Letter we report the results of an investigation into the magnetic properties of a system with random uniaxial anisotropy. This system was originally proposed by Harris, Plischke, and Zuckermann[1] as a model for amorphous magnetic materials. Using a mean-field approach they predicted that for sufficiently small anisotropy this system would undergo a transition from its high-temperature paramagnetic phase to a low-temperature ferromagnetic phase at a Curie temperature considerably lower than that of the magnet with no random uniaxial anisotropy. More recently Harris and Zobin[2] have considered the possibility that spin-glass–like behavior might be induced by the anisotropy. Again using mean-field arguments they generated a phase diagram containing both a spin-glass and a ferromagnetic phase. Chen and Lubensky[3] have derived an effective free energy density, by assuming the magnetization to be zero for sufficiently large anisotropy, which has the same form as that of the random-bond spin-glass model with Ising symmetry. A phase with a remanent magnetization was found in numerical simulations by Chi and Alben.[4]

The critical properties of the paramagnetic-to-ferromagnetic transition have been investigated within a $4-\epsilon$ expansion by Aharony.[5] Using renormalization-group techniques he generated recursion relations for a system with random uniaxial anisotropy in which the randomly oriented anisotropy axes have both an isotropic angular distribution and a distribution with cubic anisotropy. He found that in the case of an isotropic distribution no $O(\epsilon)$ fixed point was approached. Rather, all flows from physically realizable initial Hamiltonians were "to infinity," or, more precisely, out of the range of validity of the ϵ expansion. Such flows also occurred in the case of a cubic distribution, although not necessarily for all possible initial Hamiltonians.

We have looked further into the magnetic behavior of this system. In the case of an isotropic distribution of easy- or hard-axis directions we find that *there can be no ferromagnetism in a system having fewer than four spatial dimensions*.[6] Instead of being ferromagnetic the low-temperature phase is an Edwards-Anderson spin-glass[7] in that

$$\langle\langle \vec{M}_i \rangle\rangle_A = 0 \tag{1}$$

while

$$\langle\langle \vec{M}_i \rangle\langle \vec{M}_i \rangle\rangle_A \equiv q \neq 0, \tag{2}$$

where $\vec{M}_i$ is the magnetic moment at a site and the inner brackets correspond to a thermal average while the outer brackets, $\langle \dots \rangle_A$, denote an average over easy-axis configurations. We believe that it is the replacement of the low-temperature ferromagnetic phase by a spin-glass phase that is responsible for the runaways seen by Aharony, a possibility that has also been suggested by Chen and Lubensky.[3]

In more than four dimensions a ferromagnetic phase is allowed. A schematic phase diagram is shown in Fig. 1. The phase diagram is in the $\tilde{\Delta}$-T plane, T being temperature and $\tilde{\Delta}$ a parameter measuring the strength of the random uniaxial anisotropy to be described in more detail shortly. The ferromagnetic–spin-glass phase boundary lies along the curve $\tilde{\Delta} = O(\epsilon)$ in the $4+\epsilon$ dimensions. Thus as the dimensionality approaches four from above the ferromagnetic phase disappears by being squeezed onto the T axis. The paramagnetic-to-ferromagnetic phase transition has Gaussian exponents. In fewer than six dimensions the ferromagnetic phase has infinite longitudinal zero-field susceptibility, much like the ferromagnetic phase of the isotropic n-vector model without disorder in fewer than four dimensions. The spin-glass phase can be shown to have properties similar to those of a random-bond–induced spin-glass in a single-component, Ising-like system.[3] The critical exponents of the spin-glass–to–ferromagnetic transition can be obtained from a $4+\epsilon$ low-temperature–type expansion analogous to the $2+\epsilon$ expansion for isotropic n-vector systems in slightly more than two dimensions.[8]

We will now present a brief summary of the calculations we have carried out to support the conclusions presented above.

The model we have investigated is the continuous spin-field model studied by Aharony,[5] with the following Ginzburg-Landau-Wilson Hamiltonian:

$$\mathcal{H}=\int\{\tfrac{1}{2}[r_0|\vec{\sigma}(\vec{R})|^2+|\nabla\vec{\sigma}(\vec{R})|^2-D(\hat{n}(\vec{R})\cdot\vec{\sigma}(\vec{R}))^2]+\tfrac{1}{4}u|\vec{\sigma}(\vec{R})|^4\}\,d^dR, \qquad (3)$$

where $\hat{n}(\vec{R})$ is a unit vector with an isotropic random distribution.

The anisotropy field is "quenched in" so that averages with respect to configurations of $\hat{n}(\vec{R})$ must be taken *after* thermal averages over $\vec{\sigma}(\vec{R})$ for a given configuration of $\hat{n}(\vec{R})$'s. These latter averages can be obtained as a sum of Feynman diagrams by expanding $e^{-\mathcal{H}}$ in powers of $u|\vec{\sigma}|^4$ and $D(\hat{n}\cdot\vec{\sigma})^2$. Cumulant averages are obtained as appropriate linked-cluster sums. Averaging over the $\hat{n}$'s has the effect of joining "dangling" $\hat{n}$ lines in the linked diagram.[9] An example of this last procedure is shown pictorially in Fig. 2(a).

The destruction of the ferromagnetic state by random uniaxial anisotropy can be demonstrated in the same way as Imry and Ma showed that a random magnetic field destroys ferromagnetism in an isotropic m-vector system with $m \geq 2$ in fewer than four dimensions.[10] We assume a ferromagnetic state with $\langle\langle\vec{\sigma}\rangle\rangle_A$ in the z direction and then calculate the lowest-order contribution to $\langle\langle\sigma_x(\vec{R})\rangle\langle\sigma_x(\vec{R})\rangle\rangle_A$. The lowest-order graph is shown in Fig. 2(b), before and after averaging. The corresponding expression is

$$\int G_\perp{}^2(\vec{k})D^2M^2\,d^dk, \qquad (4)$$

where

$$M=\langle\langle\sigma_z(\vec{R})\rangle\rangle_A. \qquad (5)$$

$G_\perp(\vec{k})$ is the transverse propagator. On a diagram by diagram basis it can be shown that the fully renormalized transverse propagator is gapless in the limit of zero applied constant field to all orders in D and u.[11] If we assume that $G_\perp(k)\propto k^{-2}$ then the integral in (4) is of the form $\int k^{-1}d^dk$, which diverges at the low-k limit if $d\leq 4$.

Of course, the above is not a definite proof as other diagrams with possibly canceling divergences can be added. An alternative demonstration of the destruction of ferromagnetism in fewer than four dimensions.[10] We assume a ferrois similar to Schuster's proof of the destruction of ferromagnetism by a random field.[12] However, since the latter demonstrations involve Bogoliubov inequalities whose precise meaning is clouded by the replica tricks utilized, this also cannot be considered as ironclad proof. Nevertheless we regard our conclusion as firm given the weight of evidence in its favor.

In the limiting case $m\to\infty$ the results discussed above can be verified explicitly. We define $\Delta = D^2\langle\hat{n}_\alpha{}^2\hat{n}_\beta{}^2\rangle_A$, $\alpha\neq\beta$. If we take $\Delta, u\to m^{-1}$, then the leading-order free-energy diagrams will be Cayley trees of loops and ladders like those

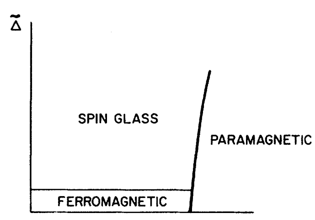

FIG. 1. Phase diagram for a magnet with isotropically distributed random uniaxial anisotropy in more than four dimensions. $\tilde{\Delta}$ is the disorder parameter defined in Eq. (11).

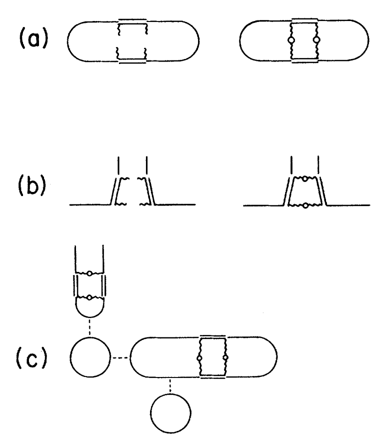

FIG. 2. (a) A free-energy diagram before and after averaging the random uniaxial anisotropy. (b) Lowest-order diagram for $\langle\sigma_x\rangle^2$ before averaging and for $\langle\langle\sigma_x\rangle^2\rangle_A$, after averaging. (c) A typical free-energy diagram, one of the leading-order contributions in the limit of $(m\to\infty)$-component systems. In the above diagrams the u vertex is denoted by a broken line, the D vertex by a double line, and $\hat{n}_\alpha$ by a wavy line.

shown in Fig. 2(c).[13] The tree and ladder sums can be collapsed into the following equation for the inverse transverse susceptibility, correct to leading order in m:

$$\Sigma + r_0 = r_0 + mu\int G_\perp(\vec{k})\, d^dk - m\Delta \int G_\perp(\vec{k})\, d^dk + uq, \tag{6}$$

with q given by the equation

$$q = M^2 + mq\Delta \int G_\perp^{\,2}(\vec{k})\, d^dk, \tag{7}$$

and $M = \langle\langle \sigma_z \rangle\rangle_A$ satisfying in zero applied field

$$(r_0 + \Sigma)M = 0. \tag{8}$$

The propagator $G_\perp(\vec{k})$ is given by

$$G_\perp^{-1}(\vec{k}) = r_0 + \Sigma + k^2. \tag{9}$$

It is not hard to demonstrate with the use of Eqs. (7)–(9) that ferromagnetism, or $M \neq 0$, is not an accessible state when $d \leq 4$, the ferromagnetic phase being replaced by a spin-glass one with $q \neq 0$ and $M = 0$. For $d > 4$ we also obtain the results described above.

The ferromagnetic and spin-glass phases in more than four dimensions can be studied in other ways. The $4+\epsilon$ expansion for the critical properties of the ferromagnetic-to-spin-glass transition starts with a fixed-length spin Hamiltonian,

$$\mathcal{H} = \int \left[-\frac{1}{2T} |\nabla \vec{S}(\vec{R})|^2 - \frac{\tilde{D}}{2T} |\hat{n}(\vec{R}) \cdot \vec{S}(\vec{R})|^2 \right] d^dR, \qquad |\hat{n}(\vec{R})|^2 = |\vec{S}(\vec{R})|^2 = 1, \tag{10}$$

assumes complete spin alignment in the z direction, and expands with respect to fluctuations about it. Recursion relations are developed for $T(l)$ and $\Delta(l)$, where

$$\tilde{\Delta} = \tilde{\Delta}(0) = \tilde{D}^2 \langle \hat{n}_\alpha^{\,2} \hat{n}_\beta^{\,2} \rangle_A, \quad \alpha \neq \beta. \tag{11}$$

The recursion relations are

$$dT(l)/dl = (2-d)T(l) + [(m-2)/8\pi^2]T(l)[\tilde{\Delta}(l) + T(l)] + O(T^3, T^2\tilde{\Delta}, \tilde{\Delta}T^2), \tag{12}$$

$$d\tilde{\Delta}(l)/dl = (4-d)\tilde{\Delta}(l) + [\tilde{\Delta}(l)/8\pi^2]\{(m-2)[\tilde{\Delta}(l) + T(l)] - T(l)\} + O(\tilde{\Delta}^3, T^2\tilde{\Delta}, \tilde{\Delta}T^2). \tag{13}$$

$\tilde{\Delta}(l)$ has an $O(\epsilon)$ fixed point in $4+\epsilon$ dimensions, yielding for the critical exponent ν the lowest-order result, $\nu = 1/\epsilon$. For η we obtain $\eta = \epsilon/(m-2)$. The results for ν and η are the same as those obtained in $2+\epsilon$ expansion for the σ^4 model. We find that hyperscaling is violated in the ferromagnetic-to-spin-glass transition. To first order in $\epsilon = d-4$, the hyperscaling relation $d\nu = 2-\alpha$ is replaced by $(d-2)\nu = 2-\alpha$. This is the same relation as that obtained for the magnetic transition in the random field model.[14]

The phase transition between the paramagnetic and ferromagnetic phases in more than four dimensions can be studied using the "soft-spin" Hamiltonian (3). The fixed point governing the transition is the Gaussian fixed point $u_r = \Delta_r = 0$, u_r and Δ_r being the fully renormalized quartic coefficient and randomness parameter. We have here the interesting case of a transition that does not exist unless its exponents are Gaussian. The anomalous longitudinal susceptibility of the ferromagnetic phase in fewer than six dimensions is the result of the massless transverse fluctuations. Working in $6-\bar{\epsilon}$ dimensions one can carry out an analysis similar to Nelson's investigation of an isotropic n-vector system in fewer than four dimensions.[15] We find that the sum of the most divergent diagrams for χ_L of the random uniaxial anisotropy model in the ordered phase is identical to that for the specific heat of an $(m-1)$-component random *field* model in the *disordered* phase. On the critical isotherm the longitudinal susceptibility behaves as $h^{-(\delta-1)/\delta}$, while as $h \to 0$ at fixed $T < T_c$, χ_L goes as $h^{-\bar{\epsilon}/2}$. For $d=6$, χ_L diverges as $\ln h$. In the disordered phase all the exponents are found to be Gaussian for $d>4$ in agreement with Aharony.[5] The behavior of the longitudinal susceptibility in the ordered phase is the only indication that the upper critical dimension for this model is 6.

Possible experimental systems exhibiting the type of spin-glass transition discussed in this Letter include the DyCu, TbAg, DyNi, and DyAl alloy systems.[16]

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[(a)]Permanent address: Department of Physics, Harvard University, Cambridge, Mass. 02138.
[(b)]Present address: Physics Department, Tufts University, Medford, Mass. 02155.
[1]R. Harris, M. Plischke, and M. J. Zuckermann, Phys. Rev. Lett. 31, 160 (1973). See also R. W. Cochrane, R. Harris, M. Plischke, D. Zobin, and M. J. Zuckermann, J. Phys. F 5, 763 (1975).
[2]R. Harris and D. Zobin, in *Magnetism and Magnetic Materials—1975*, AIP Conference Proceedings No. 29, edited by J. J. Becker, G. H. Lander, and J. J. Rhyne (American Institute of Physics, New York, 1976), p. 156.
[3]J. H. Chen and T. C. Lubensky, Phys. Rev. B 16, 2106 (1977).
[4]M. C. Chi and R. Alben, J. Appl. Phys. 48, 2987 (1977).
[5]A. Aharony, Phys. Rev. B 12, 1038 (1975).
[6]This result has also been suggested by U. Krey, to be published.
[7]S. F. Edwards and P. W. Anderson, J. Phys. F 5, 965 (1975).
[8]D. R. Nelson and R. A. Pelcovits, Phys. Rev. B 16, 2191 (1977).
[9]A. B. Harris and T. C. Lubensky, Phys. Rev. Lett. 33, 1540 (1974).
[10]Y. Imry and S.-K. Ma, Phys. Rev. Lett. 35, 1399 (1975).
[11]R. A. Pelcovits, E. Pytte, and J. Rudnick, to be published.
[12]H. G. Schuster, Phys. Lett. 60A, 89 (1977).
[13]S.-K. Ma, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1976), Vol. 6, p. 250.
[14]G. Grinstein, Phys. Rev. Lett. 37, 944 (1976).
[15]D. R. Nelson, Phys. Rev. B 13, 2208 (1976).
[16]J. M. D. Coey, in Proceedings of the Twenty-Third Annual Conference on Magnetism and Magnetic Materials, Minneapolis, Minnesota, 8–11 November 1977 (to be published), and other references listed in this review.

Two-Dimensional Electrical Conductivity in Quench-Condensed Metal Films

R. C. Dynes, J. P. Garno, and J. M. Rowell
Bell Laboratories, Murray Hill, New Jersey 07974
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The resistance of ultrathin films condensed on low-temperature substrates has been measured. All of the materials studied (Au, Cu, Al, Sn, and Pb) show a transition from activated to metallic behavior as the resistance is reduced below 30 000 Ω/□. This result gives strong evidence in support of the concept of a minimum two-dimensional conductivity. In Pb films with resistance > 30 000 Ω/□, the onset of superconductivity is marked by an increase in activation energy.

There is growing evidence[1,2] that as the resistance of a two-dimensional electron system is increased, for example by reducing the electron density in inversion layers[3] or by changing topology in thin metal films,[4] metallic conduction cannot persist above a resistance of $\sim 0.12e^2/\hbar = 30\,000\ \Omega/\square$. This limit occurs at the Ioffe-Regel[5] condition, when the electronic mean free path is equal to the Fermi wavelength. Above this limit conduction becomes activated. It is clear that such a high resistance cannot be reached in a uniform metal film, where the Fermi wavelength is comparable to the lattice spacing, as even a poor conductivity of approximately $10^4\ (\Omega\ \mathrm{cm})^{-1}$ would require a film thickness of less than an interatomic spacing. Hence when films reach the resistance of 30 000 Ω/□, they must be nonuniform, consisting of islands connected by narrow constrictions or by tunnel barriers. There has been considerable discussion concerning the energy barriers determining this activated transport. The splitting of energy levels within each island or constriction due to the size effect may give rise to the activation energy,[1,2] or the charging energy of such small particles may be dominant.[6]

In this Letter we present a study of films made by evaporation of Pb, Sn, Au, Al, and Cu onto a substrate held near 4.2 K. Such quench-condensed films show conductivity at a lower average thickness than films of the same materials made at higher temperatures. Further, *in situ* measurement of the resistance versus temperature has allowed us to increase the thickness of the films progressively until activated conductivity is no longer observed. This transition from activated to metallic behavior occurs near a resistance of 30 000 Ω/□. We further observe that in Sn films the resistance varies as $\exp(T^{-1})$, whereas in Au and Cu it varies as $\exp(T^{-1/2})$. In the latter cases the extrapolated high-temperature resistance is close to 30 000 Ω/□. In Pb films we observe tunneling between superconducting islands in the activated regime and supercurrents due to Joseph-