Coexistence of Spin-Glass and Ferromagnetic Orderings

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The phase diagram of the infinite-range model of spin-glasses exhibits two mixed phases. In these mixed phases, ferromagnetism and spin-glass order coexist, due to freezing of the transverse degrees of freedom or replica symmetry breaking. This may help to interpret a number of recent experimental findings, e.g., in *Au*Fe.

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Many disordered magnetic materials have been systematically investigated as a function of the concentration of the constituents. In the phase diagrams of these materials, one often finds a spin-glass phase for some range of concentration and a phase with long-range order (ferromagnetic or antiferromagnetic) for other concentrations.[1-23] Recently, a number of experiments have been performed to study the border region between the spin-glass and ferromagnetic phases, and many interesting data are now available. Unfortunately these data are most often interpreted, qualitatively[2,10,16] and even quantitatively,[4,17,20,24] with the help of a theory[25] which, though of great merit in its days, is now obsolete (it has been proven to be incorrect,[26] at low temperatures, as a solution of its model). Moreover, these theoretical predictions are inadequate because they are concerned with Ising spins, whereas experimental spins are Heisenberg-like (isotropic vector spins). The purpose of this Letter is to draw attention to recent studies,[27-31] which should provide a more satisfactory theoretical basis, and to present new results concerning the nature of two mixed phases, with both spin-glass and ferromagnetic characters. The existence of such intermediate phases may help explain a number of phenomena already observed experimentally, but hitherto ambiguously interpreted.

In the spirit of a mean-field theory, we consider the famous infinite-range model[25] for N classical vector spins $\vec{S}_i$. Each $\vec{S}_i$ has m components $S_{i\mu}$ ($\mu = 1, \ldots, m$) satisfying, for convenience, the following normalization condition:

$$\sum_{\mu=1}^{m} S_{i\mu}^2 = m. \tag{1}$$

They interact via independent random interactions J_{ij} distributed according to the following law:

$$P(J_{ij}) = \left(\frac{N}{2\pi}\right)^{1/2} \exp\left[-\frac{N}{2}\left(J_{ij} - \frac{J_0}{N}\right)^2\right] \tag{2}$$

so that $\langle J_{ij}\rangle_b = J_0/N$ and $\langle J_{ij}^2\rangle_b = 1/N$, where $\langle\cdots\rangle_b$ denotes an average over the bond disorder, that is, over $P(J_{ij})$. In the presence of an external magnetic field H applied along the $\mu = 1$ direction, the Hamiltonian of the model reads

$$\mathcal{H} = -\sum_{(ij)} J_{ij} \sum_{\mu} S_{i\mu} S_{j\mu} - H \sum_{i} S_{i1}, \tag{3}$$

where the sum over (ij) denotes a summation over the $N(N-1)/2$ distinct pairs of sites.

In the limit of large and positive J_0, the interactions are mainly ferromagnetic and the system exhibits a ferromagnetic phase. For $J_0 = 0$, the interactions are random in sign and the ordering is of spin-glass type. The border region between ferromagnetic and spin-glass phases occurs for $J_0 \simeq 1$. However, it has been shown[29] how the properties of the model for $J_0 \neq 0$ can be simply derived from the case $J_0 = 0$, which we therefore first consider.

In the thermodynamic limit $N \to \infty$, with use of the celebrated replica trick,[25] the free energy per spin can be determined by (so long as no replica symmetry breaking occurs),

$$f = -\frac{1}{4T}\left[m + \sum_{\mu}(q_\mu^2 - s_\mu^2)\right] - \frac{T}{(2\pi)^{m/2}} \int\cdots\int \left(\prod_{\mu} dt_\mu\right) \exp\left(-\tfrac{1}{2}\sum_{\mu} t_\mu^2\right)$$
$$\times \ln \mathrm{Tr} \exp\left(\frac{1}{T}\sum_{\mu} t_\mu q_\mu^{1/2} S_\mu + \frac{1}{2T^2}\sum_{\mu}(s_\mu - q_\mu) S_\mu^2 + \frac{1}{T} H S_1\right). \tag{4}$$

The q_μ and s_μ (Ref. 32) are variational parameters, with the following physical significance: $q_\mu\langle\langle S_\mu\rangle_{th}^2\rangle_b$ is the Edwards-Anderson (EA) order parameter, $s_\mu=\langle\langle S_\mu^2\rangle_b\rangle_{th}-1$ is a quadrupolar deformation parameter; and the $\langle\cdots\rangle_{th}$ means thermal averaging. The relation (1) implies the constraint:

$$\sum_\mu s_\mu = 0. \tag{5}$$

In the Ising case ($m=1$), there is no need to introduce the parameter s_μ and expression (4) for f simplifies to a well known form.[25]

We have performed expansions in powers of H, q_μ, and s_μ up to the fourth order, which is equivalent to a high-temperature expansion for f up to the seventh order in $1/T$. This provides information in the region $T\simeq 1$, H small of the (H, T) phase diagram. We have also made calculations for low temperatures and high fields. The resulting phase diagram, shown in Fig. 1, is now discussed.

In finite field, two transition lines come out of the zero-field transition point at $T_c=1$. Line (a) separates the high-temperature paramagnetic phase from a spin-glass phase, characterized by a freezing of the spin components which are transverse with respect to the applied field.[31] The order parameter for this transition is the transverse part of the EA order parameter, namely $q_t=q_\mu$ with $\mu\neq 1$. Above the transition, q_t vanishes whereas q_1 remains finite, due to the polarization of the spins by the applied field.

In the framework of the infinite-range model for m-component spins, the position of line (a), defined by the equation $H=H_1(T)$, can be found in two limits:

$$H_1^2 \simeq 4\left(\frac{m+2}{m+4}\right)(1-T), \quad \text{for } T\lesssim 1, \tag{6}$$

$$T \simeq K_1(m)\exp\left(-\frac{H_1^2}{2m}\right), \quad \text{for } T\simeq 0. \tag{7}$$

On physical grounds, it is clear that this transition does not exist in the Ising case, $m=1$, where no transverse components are left.

Line (b) is the instability line, corresponding to spontaneous breaking of the replica symmetry, which was first found by de Almeida and Thouless,[26] for the case $m=1$. For general m, the position of line (b), defined by the equation $H=H_2(T)$, is obtained in the two limits as:

$$H_2^2 \simeq \frac{4}{m+2}(1-T)^3, \quad \text{for } T\lesssim 1, \tag{8}$$

$$T \simeq K_2(m)\exp\left(-\frac{H_2^2}{2m}\right), \quad \text{for } T\simeq 0. \tag{9}$$

In the limit $m\to 1$, line (b) tends toward its Ising position, which is known with high precision[28,30,32] both analytically and numerically. Detailed predictions have been presented for the physical properties of the low-temperature spin-glass phase in the Ising case,[27] such as the existence of a plateau for the magnetization as a function of temperature for a constant applied field (projection hypothesis[28]). Numerical tests have been performed[33] to check these predictions and others are under way. For the general case of vector spins, much less is known but the persistence of such a plateau in the low-temperature spin-glass phase is at least consistent with the results[34] for $m\to\infty$.

We now turn to the case $J_0\neq 0$ and to the transition region from spin-glass to ferromagnetic order, which is our main interest here. The previous results allow us to draw the phase diagram (T, J_0), in zero applied field, shown in Fig. 2. The existence of paramagnetic (P), spin-glass (SG), and ferromagnetic (F) phases were previously recognized.[25-31,33-36] The novel features of the phase diagram, i.e., the mixed phases M_1 and M_2, have received only scant attention up to now.[29,31,35]

The mixed phase M_1 is characterized by the coexistence of a spontaneous magnetization (ferromagnetic order) and a spin-glass ordering of the transverse components of the spins. In a three-dimensional phase diagram (H, T, J_0), the transition line of Fig. 2 between the phases F and M_1 belongs to a transition surface which also contains line (a) of Fig. 1. In the infinite-range

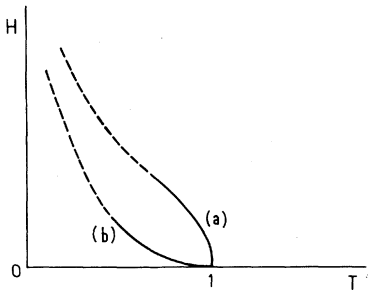

FIG. 1. Phase diagram of the infinite-range model for classical vector spins (H magnetic field, T temperature). Line (a) corresponds to the freezing of the transverse degrees of freedom, line (b) to the de Almeida-Thouless line (see text).

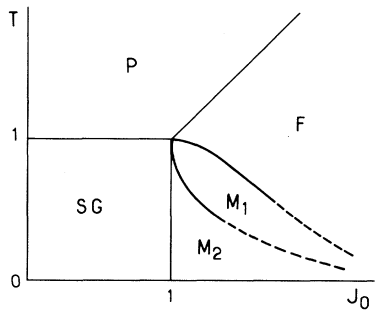

FIG. 2. Phase diagram in zero applied field (J_0 is the mean ferromagnetic interaction). Phases: paramagnetic (P), ferromagnetic (F), spin glass (SG), and mixed (M_1 and M_2).

model, the position of the F-M_1 transition line $T_1(J_0)$, in the vicinity of the multicritical point, is given by

$$1 - T_1 \simeq \frac{1}{2}\,\frac{m+4}{m+2}\,(J_0 - 1)^2, \quad \text{for } J_0 \gtrsim 1. \tag{10}$$

For J_0 large, T_1 vanishes asymptotically.

The mixed phase M_2 has the same coexistence of orderings as phase M_1 and in addition has the spontaneous breaking of replica symmetry. For Ising spins ($m = 1$), phase M_1 does not exist and there is only one mixed phase, the continuation of phase M_2. For general m, the transition line of Fig. 2 between the mixed phases M_1 and M_2, defined by $T = T_2(J_0)$, behaves in the vicinity of the multicritical point according to

$$1 - T_2 \simeq \left(\frac{m+2}{3}\right)^{1/2} (J_0 - 1)^{1/2}, \quad \text{for } J_0 \gtrsim 1. \tag{11}$$

This transition line belongs to a transition surface of the (H, T, J_0) phase diagram [it also contains line (b) of Fig. 1] which appears to lie below the transverse freezing surface.

Finally, the transition line between the spin-glass phase and the mixed phase M_2 has been drawn vertical in Fig. 2. This is a consequence of the conjecture[27-31] that the low-temperature susceptibility χ is a constant in the case $J_0 = 0$. The equation of this line is actually

$$J_0(T) = \chi^{-1}(J_0 = 0). \tag{12}$$

All the quantitative results presented above have been obtained in the framework of the infinite-range model, and they may be considered physically as mean-field theory predictions. In a real disordered material, the role of the parameter J_0 will be played by a concentration c of magnetic atoms, e.g., Fe atoms in AuFe alloys. The shapes of the transition lines in such a (T, c) phase diagram may be quite different from the predictions of Fig. 2, but it is reasonable to expect that the number of phases and their relative positions will be preserved.

One archetypal material, which has been investigated by several groups[2,3,14,15,22] is the alloy AuFe, for which ferromagnetic ordering sets in when the Fe concentration c becomes larger than a critical value $c_0 \simeq 15\%$. For a given $c \gtrsim c_0$, when the temperature is decreased, a first transition, to ferromagnetism, is clearly observed. At lower temperatures, a second transition has been reported by at least two groups,[3,5] but at temperatures which differ appreciably; this second transition has been interpreted as a disappearance of ferromagnetism. However, in a recent careful neutron analysis,[15] a total of three transitions have been identified, and moreover it has been found that the ferromagnetic ordering persists down to the lowest temperatures. These results are in striking agreement with the predictions of Fig. 2 for $J_0 \gtrsim 1$. Additional evidence for a F-M_1 phase transition in AuFe has been obtained from recent transport measurements.[22]

In conclusion, we have proved that long-range ferromagnetic order can indeed coexist with spin-glass order; we have demonstrated this coexistence in our model. Moreover, we have identified two types of such mixed phases. To avoid confusion, it should be stressed that this coexistence has nothing to do with a spatially segregated coexistence, such as one infinite ferromagnetic cluster decoupled from finite superparamagnetic clusters, which has been sometimes discussed in the literature. In our analysis, the coexistence takes place everywhere in the material.

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[1]T. R. McGuire, T. Mizoguchi, R. J. Gambino, and S. Kirkpatrick, J. Appl. Phys. 49, 1689 (1978).

[2]G. J. Nieuwenhuys, B. H. Verbeek, and J. A. Mydosh, J. Appl. Phys. 50, 1685 (1979), and earlier references therein.

[3]B. R. Coles, B. V. B. Sarkissian, and R. H. Taylor, Philos. Mag. B 37, 489 (1978).

[4]B. H. Verbeek, G. J. Nieuwenhuys, H. Stocker, and J. A. Mydosh, Phys. Rev. Lett. 40, 586 (1978).

[5]B. H. Verbeek and J. A. Mydosh, J. Phys. F 8, L109 (1978).

[6]H. v. Löhneysen, J. L. Tholence, and F. Steglich, Z. Phys. B 29, 319 (1978).
[7]K. Adachi, R. Imura, M. Matsui, and H. Sawamato, J. Phys. Soc. Jpn. 44, 114 (1978).
[8]J. Dumas, C. Schlenker, G. Chouteau, M. Nasr Eddine, and F. Sayetat, J. Appl. Phys. 49, 1622 (1978).
[9]D. W. Carnegie, Jr., C. J. Tranchita, and H. Claus, J. Appl. Phys. 50, 7318 (1979).
[10]H. Maletta and P. Convert, Phys. Rev. Lett. 42, 108 (1979).
[11]H. Maletta and W. Felsch, Phys. Rev. B 20, 1245 (1979).
[12]D. W. Carnegie, Jr., and H. Claus, Phys. Rev. B 20, 1280 (1979).
[13]J. A. Mydosh, G. J. Nieuwenhuys, and B. H. Verbeek, Phys. Rev. B 20, 1282 (1979).
[14]P. A. Beck, Solid State Commun. 34, 581 (1980).
[15]A. P. Murani, Solid State Commun. 34, 705 (1980).
[16]S. M. Shapiro, G. Shirane, B. H. Verbeek, G. J. Nieuwenhuys, and J. A. Mydosh, Solid State Commun. 36, 167 (1980).
[17]S. M. Bhagat, J. A. Geohegan, and H. S. Chen, Solid State Commun. 36, 1 (1980).
[18]C. R. Fincher, Jr., S. M. Shapiro, A. H. Palumbo, and R. D. Parks, Phys. Rev. Lett. 45, 474 (1980).
[19]R. R. Galazka, S. Nagata, and P. Keesom, Phys. Rev. B 22, 3344 (1980).
[20]Y. Yeshurun, M. B. Salamon, K. V. Rao, and H. S. Chen, Phys. Rev. Lett. 45, 1366 (1980).
[21]A. Berton, J. Chaussy, J. Odin, R. Rammal, J. Souletie, J. L. Tholence, R. Tournier, F. Holtzberg, and S. von Molnar, in Proceedings of the Twenty-Sixth Annual Conference on Magnetism and Magnetic Materials, Dallas, Texas, November 1980 [J. Appl. Phys. (to be published)].
[22]A. Hamzić and I. A. Campbell, J. Phys. (Paris), Lett. 42, L17 (1981), and to be published.
[23]N. Cowlam and A. M. Shamah, J. Phys. F 11, 27 (1981).
[24]G. J. Nieuwenhuys, H. Stocker, B. H. Verbeek, and J. A. Mydosh, Solid State Commun. 27, 197 (1978).
[25]D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. 35, 1792 (1975); S. Kirkpatrick and D. Sherrington, Phys. Rev. B 17, 4384 (1978).
[26]J. R. L. de Almeida and D. J. Thouless, J. Phys. A 11, 983 (1978).
[27]G. Parisi, Phys. Rep. 67, 25 (1980), and references therein.
[28]G. Parisi and G. Toulouse, J. Phys. (Paris), Lett. 41, L361 (1980).
[29]G. Toulouse, J. Phys. (Paris), Lett. 41, L447 (1980).
[30]J. Vannimenus, G. Toulouse, and G. Parisi, J. Phys. (Paris) 42, 565 (1981).
[31]G. Toulouse and M. Gabay, J. Phys. (Paris), Lett. 42, L103 (1981); M. Gabay, thesis, Université de Paris–Sud, Orsay, 1981 (unpublished).
[32]The relevance of s_μ is discussed in Ref. 31.
[33]F. T. Bantilan, Jr., and R. G. Palmer, J. Phys. F 11, 261 (1981); S. K. Ma and M. Payne, to be published.
[34]J. R. L. de Almeida, R. C. Jones, J. M. Kosterlitz, and D. J. Thouless, J. Phys. C 11, L871 (1978).
[35]G. Toulouse, J. Vannimenus, and J. M. Maillard, J. Phys. (Paris), Lett. 38, L459 (1977); J. H. Chen and T. C. Lubensky, Phys. Rev. B 16, 2106 (1977); A. J. Bray and M. A. Moore, J. Phys. C 13, 419 (1980); J. Villain, Z. Phys. B 33, 31 (1979).
[36]D. Sherrington and B. W. Southern, J. Phys. F 5, L49 (1975).

Photoemission from Ag-O-Cs

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X-ray photoelectron spectra of Ag, O, and Cs and quantum yield distributions from surfaces prepared by evaporating Cs onto Ag oxidized in ultrahigh vacuum have been measured. The structure of this surface determined from these measurements consists of silver particles dispersed in a matrix of Cs_2O with a layer of $Cs_{11}O_3$ on top. The threshold at 1.2 μm is due to a lowering of the surface barrier by the $Cs_{11}O_3$ and the yield in the visible and near infrared is explained as due to the excitation of Mie or optical resonance absorption in the silver particles.

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The Ag-O-Cs or *S*-1 photocathode is unique among photoemissive materials with useful sensitivity to visible light. It was the first, and for many years the only, cathode available for practical applications.[1] Because of its useful response in the visible and near-infrared region of the spectrum, it continues to find widespread use in many photoelectronic applications. In spite of this no explanation of the observed quantum yield in this spectral region has been previously