Order-Disorder Transition in an External Field in Random Ferromagnets and Nematic Elastomers

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We study theoretically the effect of external field on random anisotropy ferromagnets and nematic elastomers. The system evolves on increasing field from the correlated spin glass state to the aligned ferromagnetic state via a first order phase transition at a threshold field accompanied by a significant jump of the average magnetization. The mean magnetization growth is due to reorientation, rather than growth of individual domains. We discuss the additional hardening mechanisms in nematic elastomers that increase the threshold of the transition observed experimentally and lead to the "fossil" domain walls left in the aligned system. [S0031-9007(97)04747-9]

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The presence of quenched random disorder is implicit to many physical systems, as different as random ferromagnets, vortex lattices in type-II superconductors, and liquid crystalline elastomers. It is well understood theoretically that even a weak random field of quenched defects breaks the long-range order in less than four dimensions [1–3]. This result was first obtained for vortex arrays by Larkin, and later generalized by Imry and Ma [1], who showed that in 3D the displacement correlation function grows linearly with distance. A modern analysis, based on the concept of replica symmetry breaking, revealed that at long distances a quasi-long-range order in vortices arrays pertains, although the genuine long-range order is indeed broken by random fields [3].

The influence of homogeneous external fields on a system with random disorder was investigated theoretically by Cullen [4] and Chudnovsky *et al.* [5]. They applied the random anisotropy model to describe amorphous ferromagnets in a magnetic field. It was confirmed [5] that a ferromagnet with weak random anisotropy in the absence of an external field is in the correlated spin glass (CSG) state, where the spin orientation correlations decrease exponentially with a coherence distance much bigger than an atomic scale a. Already rather weak magnetic fields nearly align the spins and give rise to the regime of a ferromagnet with wandering axis (FWA), where spins are still able to deviate slightly from the magnetic field direction; strong fields align all spins in the same direction. The correctness of this picture was proven experimentally (see, for example, the review [6]). In all theoretical models the presence of long-range order in a strong field has been assumed *a priori.* The actual process of imposing the long-range order by an external field is not described theoretically and a more rigorous analysis is needed.

The macroscopic ordering of nematic elastomers under a uniaxial stress has been observed experimentally [7–10]. With no stress applied, the ground state is polydomain, analogous to the CSG in random magnetic systems. The role of random impurities here is played by the network cross-links, which impose a local orientation on the surrounding director field. Cross-links always possess a degree of anisotropy and their positions and orientations are random and topologically fixed during the network formation. There is a clear transition, at a critical value of stress, into a macroscopically aligned state, which is called monodomain in spite of the "fossil" remains of domain walls still present in the system even at high stress. Direct observations of the polydomain-monodomain transition under external stress, now available in many different nematic elastomers, and the absence of the apparent critical transition in random ferromagnets, further motivate our study. We argue that the magnetic transition should happen at rather small fields, perhaps below the investigated range, whereas in elastomers an additional rubber-elastic effect causes the threshold stress to be significantly greater.

In this Letter we consider a random-anisotropy ferromagnet in external magnetic field. The approach is equally applicable to nematic elastomers, where the source of orientational disorder is thought to be the network cross-links. The combination of the replica trick and the Gaussian variational method developed for random manifolds [11] is used to obtain a nontrivial replica-symmetric solution which corresponds to the macroscopically ordered (ferromagnetic) phase. We carry out the analysis of stability, which shows the presence of the phase transition at a critical value of the field, between the low-field replica symmetry-broken CSG state and the high-field FWA state with a tail of long-range order. We do not address the physics of the replica symmetry-broken state and the dynamic properties of the system.

The model.—We consider a ferromagnet with randomly distributed impurities, which locally impose an easy spin orientation [12]. The Hamiltonian of such system can be written as

$$H = \int d^3r \left[\frac{\alpha}{2} (\nabla \mathbf{M})^2 - \frac{\gamma}{2} \rho(r) (\mathbf{k}\mathbf{M})^2 - h(\mathbf{n}_0 \mathbf{M}) \right], \tag{1}$$

where α and γ are the exchange and random anisotropy constants, $\mathbf{M}$ the local magnetization, and h the amplitude of the external (magnetic) field directed along the unit vector $\mathbf{n}_0$. $\rho(r) = \sum \delta(r - r_i)$ is the continuum density of impurities, which impose a completely random orientation of an easy anisotropy axis $\mathbf{k}$.

It is convenient to take the magnetization and random anisotropy axes both confined to the x-y plane, though being dependent on all three spatial coordinates (3D XY model). This choice ($n = 2, d = 3$) does not seem to affect the results qualitatively [5] but substantially simplifies the analysis. The local coarse-grained magnetization is changing the direction slowly, so that $\mathbf{M} = M\{\cos\theta, \sin\theta\}$ is parametrized by a single angle with $M = \text{const}$.

We assume the defects to be distributed with the Gaussian probability:

$$P[\rho] \simeq \exp\left\{-\int d^3r\, \frac{\rho^2}{2\rho_0}\right\}, \tag{2}$$

where ρ_0 is the mean density of impurities, and $P[\mathbf{k}] = \frac{1}{2\pi}$, signifying an arbitrary orientation of random anisotropy axis in the x-y plane.

To calculate averages over the quenched disorder we apply the replica trick [11,13]. After averaging over the random distributions of $\rho(r)$ and $\mathbf{k}$, the replica Hamiltonian takes the form

$$H_{\text{repl}} = \sum_{a,b=1}^{m} \int d^3r \left[\delta_{ab}\left(\frac{\hat{\alpha}}{2}(\nabla\theta_a)^2 - \hat{h}\,\cos\theta_a\right) - \frac{\Gamma}{4}\cos(2\theta_a - 2\theta_b)\right], \tag{3}$$

where a, b are the replica indices and m the number of replicas. The coupling parameter $\Gamma = \gamma^2\rho_0 M^4/16kT$ and constants $\hat{\alpha} = \alpha M^2$, $\hat{h} = hM$ incorporate the magnetization amplitude. This type of Hamiltonian with the periodic replica interaction has been studied before [3] to examine the glasslike disorder at zero field.

Following [3,11], we approximate the replica Hamiltonian using the Gaussian variational method. The trial Hamiltonian H_0 in Fourier space has the form

$$H_0 = \frac{1}{2}\sum_{a,b=1}^{m}\int_{q_{\min}}^{q_{\max}} \frac{d^3q}{(2\pi)^3}\, G_{ab}^{-1}(q)\theta_a(q)\theta_b(-q), \tag{4}$$

where G_{ab}^{-1} is an $m \times m$ matrix of variational parameters, $q_{\min} = 2\pi/L$ is the lower and $q_{\max} = 2\pi/a$ the upper cutoffs (L being the system size and a the atomic scale). G_{ab} describes correlations between Fourier components of magnetization on different replicas: in our notation $G_{ab} = (1/kT)\langle\theta_a(q)\theta_b(-q)\rangle$. This matrix of variational parameters G_{ab} is chosen in the convenient form: $G_{ab}^{-1} = \delta_{ab}(\hat{\alpha}q^2 + D_a) - \sigma_{ab}$. Two separate sets of parameters D_a and σ_{ab} are necessary to fit both diagonal (external field) and off-diagonal (replica interaction) parts of the variational free energy $F_{\text{var}} = -kT\ln Z_0 + \langle H_{\text{repl}} - H_0\rangle_{H_0}$. Minimization of F_{var} with respect to parameters D_a and σ_{ab} provides the stationary conditions

$$D_a = \hat{h}e^{-\frac{kT}{2}\int_q G_{aa}(q)} + \sigma_{aa} + \Gamma\sum_{c\neq a} e^{-4kT\int_q \frac{1}{\hat{\alpha}q^2 + D_c}}, \tag{5}$$

$$\sigma_{a\neq b} = 2\Gamma e^{-2kT\int_q (G_{aa} + G_{bb} - 2G_{ab})}. \tag{6}$$

We examine the class of replica symmetric solutions of Eqs. (5) and (6), i.e., with all $\sigma_{ab} = \sigma$, $D_a = D$, $G_{ab}(q) = G(q)$, and their stability. Equations (5) and (6) cannot be solved analytically, but reduce to transcendental equations for D and σ. G^{-1} can be inverted:

$$G(q) = \frac{1}{kT}\langle|\theta_a(q)|^2\rangle = \frac{1}{\hat{\alpha}q^2 + D} + \frac{\sigma}{(\hat{\alpha}q^2 + D)^2}. \tag{7}$$

One can see how the external field influences the system. The diagonal variational parameter D plays the role of mass and suppresses both the thermal and the quenched disorder (first and second terms in $G(q)$, respectively). The nonzero mass penalizes long wavelength fluctuations and limits their amplitude. This effect will become stronger with increasing external field and more short wavelength fluctuations will be suppressed. When this happens on the length scale of characteristic domain size, one can expect the onset of a macroscopically ordered (FWA) phase.

The transition.—First of all, Eqs. (5) and (6) always have a trivial solution:

$$\sigma = 2\Gamma e^{-4kT\int_q \frac{1}{\hat{\alpha}q^2}} = 2\Gamma\omega; \qquad D = 0, \tag{8}$$

where $\omega = \exp\{-2kTq_{\max}/\pi^2\hat{\alpha}\}$ is a constant of the order of e^{-1} (if the natural estimate of the exchange coupling, $\alpha M^2 \sim kT_c/a$, with T_c the temperature of magnetic ordering, is valid). This trivial solution leads to the correlation function $B(x) = \langle[\theta_a(x) - \theta_a(0)]^2\rangle \simeq \text{const} - (x/\xi_0)\ln(L/x)$. Here $\xi_0 = \hat{\alpha}^2\pi^2/kT\Gamma\omega = 16\pi^2\alpha^2/\omega\rho_0\gamma^2$ is the correlation distance, or the characteristic domain size of the CSG system in the absence of the external field. This long-range correlator is equivalent to Larkin's result [1]. This solution is known to be unstable with respect to the replica symmetry breaking. It can be checked from the eigenvalue λ of the replicon mode [3]:

$$\lambda = 1 - \frac{kT}{2}\Gamma e^{-4kT\int_q \frac{1}{\hat{\alpha}q^2}}\int_q \frac{1}{(\hat{\alpha}q^2)^2} \simeq 1 - \frac{L}{8\pi\xi_0}. \tag{9}$$

For system sizes $L \gg \xi_0$ the eigenvalue is negative and the trivial replica-symmetric solution is unstable with respect to perturbations. As a result, the system exhibits a glasslike disorder at large length scales, as in other analogous cases [3].

The trivial solution (8) does not take into account the external field and can be obtained directly from the Hamiltonian (3) with $h = 0$. To be more precise, in this

case D will be exponentially small: $D \sim \exp\{-L/\xi_0\}$ and will vanish in a large system.

To account for the influence of external field one needs to find a solution with nonzero parameter D. In the case of a very weak field ($D \ll \hat{\alpha} q_{\min}^2$) we have for D:

$$D \simeq \hat{h}\omega^{1/8} \exp\left\{-\frac{Ł}{4\pi\xi_0}\right\}. \tag{10}$$

Within the accuracy $\sim D/(\hat{\alpha} q_{\min}^2)$ the values of σ and λ are not influenced by this weak field. So, such a field is unable to make the replica-symmetric solution stable and the system remains in a glassy state.

To consider the case of stronger fields, let us now suggest that $\hat{\alpha} q_{\min}^2 \ll D \ll \hat{\alpha} q_{\max}^2$, i.e., fluctuations with the wave vector $q < q_c \equiv (D/\hat{\alpha})^{1/2}$ are now suppressed by the external field, although the shorter wavelength fluctuations remain unchanged. If we discard the terms of the order of $(q_{\min}/q_c)^2$ and $(q_c/q_{\max})^2$, Eqs. (5) and (6) decouple and the value of σ in (6) is unaffected by the external field. We obtain an approximate equation for D from Eq. (5):

$$D = \hat{h}\omega^{1/8} \exp\left\{-\frac{\pi}{8}\sqrt{\frac{\hat{\alpha}}{D\xi_0^2}}\right\}. \tag{11}$$

This equation has a solution only for the external field above the threshold h_c (see Fig. 1):

$$h_c = \left(\frac{\pi e}{16\omega^{1/16}}\right)^2 \frac{\alpha M}{\xi_0^2} \simeq 1.5 \times 10^{-6}\left(\frac{\rho_0^2\gamma^4 M}{\alpha^3}\right). \tag{12}$$

At large fields ($\hat{\alpha}\xi_0^{-2} \ll D \ll \hat{\alpha} q_{\max}^2$) the exponential in Eq. (11) can be neglected and the relation $D(\hat{h})$ becomes a linear function with the slope determined by $\omega^{1/8}$. At the same time σ remains weakly influenced by the external field: $\sigma = 2\Gamma\omega(1 + \frac{\pi}{2} kTq_c/\hat{\alpha})$. Therefore, the apparent domain size remains approximately unchanged.

This replica-symmetric solution is stable. It can be checked that the eigenvalue λ becomes positive when $h \geq h_c$ and $D \geq D_c \sim \hat{\alpha}/\xi_0^2$:

$$\lambda = 1 - \frac{kT}{2}\Gamma e^{-4kT\int_q \frac{1}{\hat{\alpha}q^2+D}} \int \frac{1}{(\hat{\alpha}q^2 + D)^2}$$
$$\simeq 1 - \frac{\omega}{32\pi}\sqrt{\frac{\hat{\alpha}}{D\xi_0^2}}. \tag{13}$$

So, when external field reaches the threshold value, it binds fluctuations with $q \geq \xi_0^{-1}$ and imposes correlations between the domains. This is illustrated by the critical value q_c being indeed of the order of ξ_0^{-1}. The domain size itself does not change during the phase transition apart from the small correction term of the order of $q_c/q_{\max}$. The evolution of the CSG state, therefore, proceeds via the reorientation of correlated regions towards the field direction, while keeping their characteristic size unchanged in the first approximation.

The average magnetization in the ordered FWA is

$$\overline{M} = \frac{M}{V}\int d^3x\langle\cos\theta(x)\rangle = Me^{-\frac{kT}{2}\int_q G(q)}$$
$$\simeq \exp\left\{-\frac{kT}{4\pi^2\hat{\alpha}}\left[q_{\max} - \frac{\pi}{2}\sqrt{\frac{D}{\hat{\alpha}}}\right]\right.$$
$$\left. - \frac{\pi}{16\xi_0}\sqrt{\frac{\hat{\alpha}}{D}}\right\}, \tag{14}$$

where $D(h)$ is given by Eq. (11) and Fig. 1. The two terms in the exponent come from thermal and random disorder, respectively. They both are of the order of unity at the transition and tend to zero with the growth of external field. It gives the small jump of the average magnetization at $h = h_c$, which then increases monotonically with the growth of h (Fig. 2). At large fields an approximate form of Eq. (14) can be used; returning to the dimensional parameters $\overline{M} \simeq M\exp\{-\frac{1}{128\pi}\rho_0\gamma^2 M^{1/2}\alpha^{-3/2}h^{-1/2}\}$.

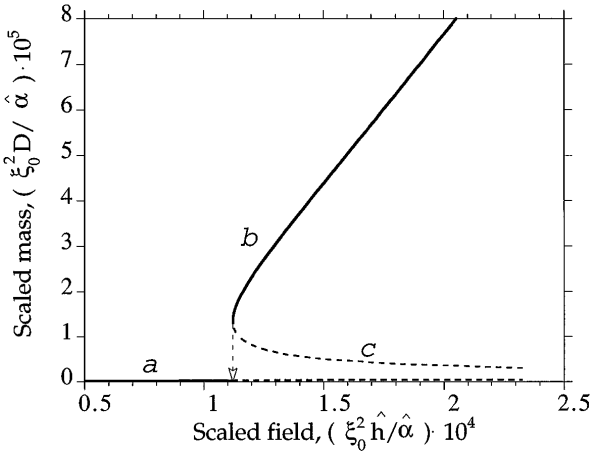

FIG. 1. The solution of Eq. (11). Variational parameter D (scaled by $\hat{\alpha}/\xi_0^2$) obtained as a function of external field (scaled by $\hat{\alpha}/\xi_0^2$). The transition from trivial (curve a) to nontrivial replica-symmetric solution (curve b) at a critical field h_c is indicated by an arrow. The dashed line c represents the unstable branch of solutions for $D(h)$.

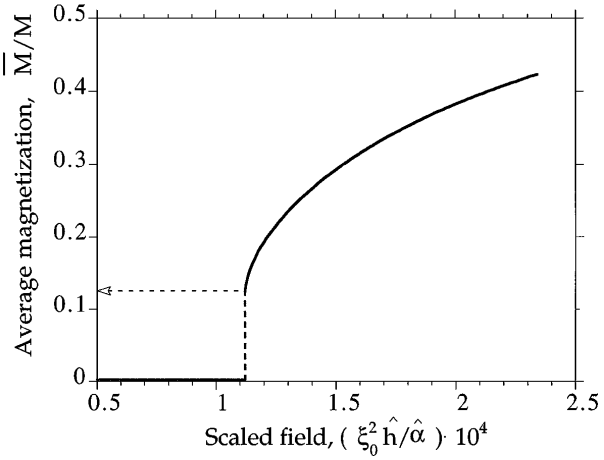

FIG. 2. The order parameter (scaled average magnetization) $\overline{M}/M$ as a function of scaled external field $\hat{h}\xi_0^2/\hat{\alpha}$, obtained using the dependence $D(h)$ from Fig. 1. In spite of a significant jump at the transition [see Eq. (14)], the order parameter behaves in a critical fashion, $\overline{M} - \Delta\overline{M} \sim (h - h_c)^{1/2}$.

Nematic elastomers.—The model is equally applicable to nematic elastomers, although there are some differences. The free energy density will have the form similar to Eq. (1) [9]. The unit director field $\mathbf{n}(r)$ replaces the magnetization, with the Frank elastic constant K instead of α. The external field couples to $\mathbf{n}$ in a quadrupolar fashion, with the corresponding term in Hamiltonian $-\tilde{h}(\mathbf{n}_0 \cdot \mathbf{n})^2$. The external field can be either mechanical (uniaxial stress) with $\tilde{h}$ the value of stress, or magnetic with $\tilde{h} = \chi_a \mathbf{h}^2$, where χ_a is the anisotropy of magnetic susceptibility. The random field here is caused by the network cross-links, which always possess a degree of anisotropy. Their positions and orientations are random and fixed during the network formation, leading to the same form of random-field term in the continuum Hamiltonian [9].

However, one should take into account the additional rubber-elastic deformation of the underlying polymer network. Elastic properties of nematic elastomers include a peculiar effect called "soft elasticity" (see the review [14])—the ability to sustain certain deformations without any energy cost due to the coupling between network deformations and director rotations. To be soft, the fluctuation should be either pure bend, pure splay, or splay-twist combinations [15]. These conditions can be satisfied only in a homogeneous director field. So, strictly speaking, a nematic elastomer is hard when in a glassy state with a highly nonuniform $\mathbf{n}$. Nonetheless the fluctuations with $q > 1/\xi_0$ can be soft inside each domain, as long as the director field at the scale ξ_0 is roughly homogeneous.

As in the CSG, the zero-field disordered elastomer does not have the pronounced interfaces between correlated regions. However, when an external stress is applied, the system would gain energy if the least possible deformation caused by external field is contained in the hard, domain interface regions. The corresponding elastic energy response, per unit volume, reads $\frac{1}{2}\mu\varepsilon^2\ell/\xi_0$; with μ the rubber modulus; ε the overall sample deformation; and ℓ the width of an interface. The elastic penalty for sustaining such an interface is estimated as $\frac{1}{2}\mu Q^2\xi_0/\ell$ per unit volume, where Q is the measure of nematic chain anisotropy of misaligned neighboring domains. Therefore, the domain wall localizes under strain: $\ell^* \sim \xi_0(Q/\varepsilon)$. The interface energy density, resulting from this balance, is $\mu Q\varepsilon$. It must be added to the external-stress term $-\tilde{h}\varepsilon$, providing a new value for the threshold: Eq. (12) will now determine the value of the offset stress $(\tilde{h}_c - \mu Q)$.

The discussed effects, accounting for the rubber network deformation, are diagonal in replicated Hamiltonian (3) and so will cause only a renormalization of the nematic Frank elastic constant K and a shift of the "mass" $D(h)$, leaving the general picture of the order-disorder phase transition unchanged. After a small jump at the threshold stress $\tilde{h}_c \sim \mu Q$, the macroscopic order parameter S will be increasing with the stress,

$$S \simeq Q \exp\left\{-\frac{\pi}{2}\,\frac{K^{1/2}}{\xi_0(\tilde{h} - \mu Q)^{1/2}}\right\}.$$

The parameter of nematic chain anisotropy Q is usually of the order 0.1–0.2 in side chain, and much greater in main-chain liquid crystal polymers. Hence the threshold of the transition is well within experimental accuracy, and has been clearly observed by many groups. The characteristic increase of the average orientation parameter $S(\tilde{h})$ has also been seen, although to resolve a predicted jump S_c one would need to ensure the equilibrium conditions (the relaxation dynamics of random elastomers is very slow).

To summarize, we obtained a discontinuous transition, driven by an external homogeneous field, between the correlated spin glass state and the ferromagnetic state with a certain degree of long-range order, and between polydomain and monodomain states of nematic elastomers. At, and past the transition the characteristic domain size ξ_0 changes only weakly with applied field and, therefore, the increase in the long-range order takes places via the reorientation of individual correlated regions, rather than through domain growth. In elastomers, due to the additional rubber-elastic energy, the domain boundaries sharpen on increasing field and are left in the system even after the complete alignment is achieved.

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