Spin-Glass State of a Randomly Diluted Granular Superconductor

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A randomly diluted lattice of Josephson tunnel junctions is used as a model to generate a replicated Landau-Ginzburg field theory describing the properties of diluted superconductors near the percolation threshold. The model predicts Meissner and Abrikosov phases and a spin-glass phase with frozen currents, a nonvanishing Edwards-Anderson order parameter, and power-law decay of correlations of the magnetic field.

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Superconductivity in disordered granular materials near a percolation threshold exhibits novel properties in the presence of an applied magnetic field. These materials consist of superconducting islands embedded in a nonsuperconducting host and are coupled by means of Josephson tunneling of Cooper pairs or proximity effect. Such systems have received considerable theoretical[1-5] and experimental attention.[1,6] Recent numerical studies have suggested that disordered systems in the presence of a sufficiently strong magnetic field may freeze into a state in which the condensate wave function exhibits spin-glass-type order among the superconducting grains.[5] In this Letter, we examine the properties of a randomly diluted granular superconductor near percolation in the low-temperature limit, and discuss the conditions under which a transition to a thermodynamic state of spin-glass superconducting order may occur. This is done from first principles by means of an $n \rightarrow 0$ replica field theory for the randomly diluted Josephson-junction network. The mean-field phase diagram of this theory (Figs. 1 and 2) has the familiar Meissner phase and an Abrikosov vortex-lattice phase. In addition we have demonstrated the existence of a "glass" phase which is the analog of a spin-glass in random magnetic systems[7]: The configurationally and thermally averaged condensate wave function $[\langle\psi\rangle_T]_c$ is zero whereas the Edwards-Anderson[7] order parameter $[|\langle\psi\rangle_T|^2]_c$ is nonzero. The glass phase is characterized by complete penetration on average of the applied magnetic field but in addition by a random distribution of frozen-in Josephson currents leading to fluctuations in the magnetic field which decay as a power law with distance. The transition from the Abrikosov to glass phase at zero temperature occurs at a magnetic field H_g corresponding to approximately one quantum of flux per typical loop of the diluted Josephson network in the Skal–Shklovskii–de Gennes[8] node-link picture of a percolating network. For a single isolated loop in which the superconducting coherence length is short compared to the loop perimeter there are the Little-Parks oscillations in the induced supercurrent as a function of applied field. Fluctuations in the sizes of these loops in a disordered network lead to damping of such oscillations.[5] Near the percolation threshold, where these fluctuations are most dramatic, frustration produced by the interaction of loops of different areas leads to the glass phase.

We consider a replicated model for a granular material in the presence of a magnetic field in which the

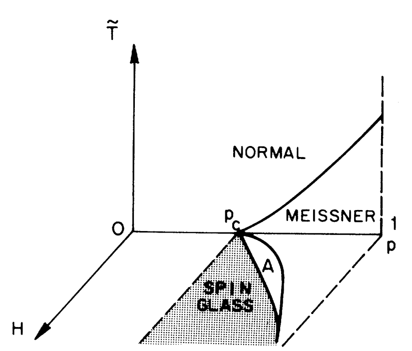

FIG. 1. Mean-field phase diagram as a function of temperature T, applied magnetic field H, and Josephson bond occupation probability p near percolation threshold p_c exhibiting normal, Meissner, glass, and Abrikosov (A) phases.

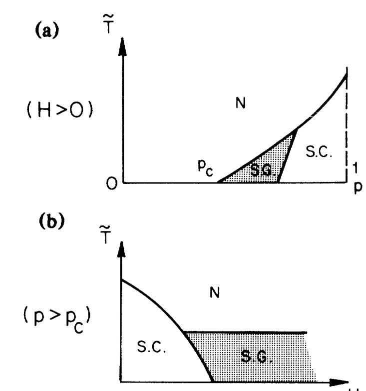

FIG. 2. (a) Phase diagram for fixed applied magnetic field H, showing spin-glass (S.G.), superconducting (S.C.), and normal (N) phases. (b) Same for fixed concentration of superconducting grains above percolation threshold p_c.

phase $\theta_{\mathbf{x}}^{\alpha}$ of the condensate wave function in a grain at site $\mathbf{x}$ interacts with a neighbor at $\mathbf{x}'$ by the Josephson coupling:

$$H^{\alpha} = -\sum_{\langle x,x'\rangle} K_{\mathbf{x},\mathbf{x}'}\cos(\theta_{\mathbf{x}}^{\alpha} - \theta_{\mathbf{x}'}^{\alpha} - A_{\mathbf{x},\mathbf{x}'}^{\alpha}), \qquad (1)$$

$$\alpha = 1, \ldots, n$$

where

$$A_{\mathbf{x},\mathbf{x}'}^{\alpha} = (2\pi/\phi_0)\int_{\mathbf{x}}^{\mathbf{x}'} \mathbf{A}^{\alpha}\cdot d\mathbf{l},$$

where $\mathbf{A}^{\alpha}$ is the vector potential, and $\phi_0 = hc/2e$ is an elementary flux quantum. Summation is taken over all nearest neighbors on the d-dimensional cubic lattice with lattice constant a, and disorder is introduced through the Josephson energy $K_{\mathbf{x},\mathbf{x}'}$ which is set equal to K or zero with respective probabilities p and $l-p$.

This model is valid for grain sizes of order of or smaller than both the bulk superconducting coherence length and the London penetration depth for the grains so that amplitude fluctuations in the condensate wave function and magnetic field within a single grain may be neglected.

The thermodynamic propeties of this system are governed by the configuration-averaged Helmholtz free energy $F = -k_{\mathrm{B}}T[\ln Z_A]_c$, where the square brackets with subscript c ($[\,]_c$) denote a quenched average over all possible realizations of intergrain Josephson coupling and

$$Z_A = \int D\theta_{\mathbf{x}}^1 \exp[-H^1/k_{\mathrm{B}}T]. \qquad (2)$$

The above average may be carried out with the aid of the replica procedure

$$[\ln Z_A]_c = \lim_{n\to 0}(1/n)\ln[Z_A^n]_c.$$

A generalization of the continuum ($a \to 0$) field theory introduced by Stephen[9] for the randomly diluted resistor network leads to an effective replica Hamiltonian H_r for the granular superconductor near a percolation threshold p_c:

$$[Z_A^n]_c = \int D\psi_{\mathbf{k}}\, e^{-H_r}, \qquad (3)$$

$$H_r = \sum_{\mathbf{k}\neq 0}\int \frac{d^dx}{a^d}\psi_{\mathbf{k}}(x)[r_k - C_1(\nabla - \frac{2\pi i}{\phi_0}\mathbf{A}^{\alpha}k_{\alpha})^2]\psi_{-\mathbf{k}}(x) + \frac{1}{6}\sum_{\substack{\mathbf{k}_1,\mathbf{k}_2\\ \neq 0}}\int\frac{d^dx}{a^d}\psi_{\mathbf{k}_1}(x)\psi_{\mathbf{k}_2}(x)\psi_{-\mathbf{k}_1-\mathbf{k}_2}(x), \quad c_1 = a^2/4d. \qquad (4)$$

Here, summation is over all nonzero n-component vectors $\mathbf{k} = (k_1, k_2, \ldots, k_n)$ in replica space with integer components k_{α}. Functional integration is over the continuum order-parameter fields $\psi_{\mathbf{k}}(x)$ which have expectation values corresponding to various moments of the XY order parameter $\exp[i\theta(x)]$:

$$\langle\psi_{\mathbf{k}}(x)\rangle = \left[\prod_{\alpha=1}^{n}\langle e^{ik_{\alpha}\theta(x)}\rangle_T\right]_c, \qquad (5)$$

where angular brackets denote an average with respect to H_r and angular brackets with subscript T ($\langle\,\rangle_T$) denote an average with respect to the original unreplicated Hamiltonian with a particular configuration of bonds. The Hamiltonian, Eq. (4), is simply the gauge-invariant generalization of that considered by Harris and Lubensky[10] in the context of the randomly diluted XY model in which

$$r_{\mathbf{k}} = (p_c - p) + b\tilde{T}k^2 + O(T^2), \quad \tilde{T} = k_{\mathrm{B}}T/K, \qquad (6)$$

in the low-temperature limit. Here, b is a positive constant which depends on the bond occupation probability p. Magnetic field fluctuations can be included in this model by the addition of the magnetic energy term and an integration over $\mathbf{A}^{\alpha}(x)$:

$$[Z^n]_c = \int D\mathbf{A}^{\alpha}[Z_A^n]_c\exp[-g\int d^dx\textstyle\sum_{\alpha}(\nabla\times\mathbf{A}^{\alpha})^2], \qquad (7)$$

where $g = (8\pi\mu_0 k_{\mathrm{B}}T)^{-1}$ and μ_0 is the bare magnetic permeability of the composite. We will refer to the models with and without electromagnetic fluctuations as model I and model II.

The vector potential is conjugate to the current density $\mathbf{J}(x)$ so that currents and current correlation functions in model I are determined by responses to the external vector potential $\mathbf{A}^{\alpha}(x)$. Thus the thermally and configurationally averaged current $[\langle J_i(x)\rangle_T]_c$ is

$$\langle J_i^{\alpha}(x)\rangle = -k_{\mathrm{B}}T(\phi_0/2\pi a)\delta\ln[Z_A^n]_c/\delta A_i^{\alpha}(x).$$

Of particular importance is the response of this current to changes in $\mathbf{A}^{\beta}$. Introducing the spatial Fourier transforms $J_i^{\alpha}(q)$ and $A_i^{\beta}(q)$ of the current and vector potential, we define $\gamma_{ij}(q) = \delta\langle J_i^{\alpha}(q)\rangle/\delta A_j^{\beta}(-q)$ which is related to fluctuations in J_i via

$$\gamma_{ij}^{\alpha\beta}(q) = \delta_{\alpha\beta}[\delta_{ij}E_c - (k_{\mathrm{B}}T)^{-1}\langle J_i(q)J_j(-q)\rangle_T]_c - (k_{\mathrm{B}}T)^{-1}(1-\delta_{\alpha\beta})[\langle J_i(q)\rangle_T\langle J_j(-q)\rangle_T]_c, \qquad (8)$$

where E_c is a condensation energy and where we used $[\langle J_i(q)\rangle_T]_c=0$ in the phases of interest to us. The helicity modulus studied by others[5] is simply $Y_{ij}=\lim_{q\to 0}[\gamma_{ij}^{11}(q)-\gamma_{ij}^{12}(q)]$. In model II, $\gamma_{ij}^{\alpha\beta}(q)$ becomes $\delta\langle J_i^\alpha(q)\rangle/\delta\langle A_j^\beta(q)\rangle$ and contains the usual local field corrections[11] to Eq. (8). It determines the **A**-field correlation function

$$D_{ij}^{\alpha\beta}(q)=\langle\delta A_i^\alpha(q)\delta A_j^\beta(-q)\rangle =k_BT\left[\frac{q^2}{4\pi\mu_0}\delta_{\alpha\beta}\delta_{ij}+\gamma_{ij}^{\alpha\beta}(q)\right]^{-1}, \tag{9}$$

where $\delta A_i^\alpha(q)=A_i^\alpha(q)-\langle A_i^\alpha(q)\rangle$ and where the right-hand side is the inverse matrix in both the $\alpha\beta$ and ij indices.

An approximate mean-field phase diagram as a function of $\tilde{T}$, p, and applied magnetic field H for this system may be obtained by consideration of the locus of points for which the quadratic part of H_r develops zero eigenvalues. In the mean-field approximation, we set $\nabla\times\mathbf{A}=\mu_0\mathbf{H}$ in each replica and obtain for each $\mathbf{k}$ the lowest eigenvalue at finite temperatures:

$$e_{\mathbf{k}}=p_c-p+b\tilde{T}k^2+(\pi\mu_0Ha^2/2d\phi_0)|S_{\mathbf{k}}|, \tag{10}$$

where $S_{\mathbf{k}}=\sum_{\alpha=1}^n k_\alpha$. For $T>0$, the lowest eigenvalues are associated with the $\mathbf{k}=(1,0,\ldots,0)$ and $(1,-1,0,\ldots,0)$ modes, respectively. At $T=0$, $e_{1,0,\ldots,0}$ is degenerate with all modes for which $|S_{\mathbf{k}}|=1$ and $e_{1,-1,0,\ldots,0}$ is degenerate with all modes for which $S_{\mathbf{k}}=0$. In this limit, the XY order parameter $\psi_{1,0,\ldots,0}$ exhibits long-range order for applied fields $H<H_g$ where

$$\mu_0H_g\xi_p^2=2d\phi_0/\pi \quad (p>p_c), \tag{11}$$

and the percolation correlation length ξ_p is given by $\xi_p/a=|p_c-p|^{-\nu}$ with $\nu=\frac{1}{2}$ in mean-field theory. However, modes for which $S_{\mathbf{k}}=0$ (of which the Edwards-Anderson[7] order parameter $\langle\psi_{1,-1,0,\ldots,0}\rangle$ or $[\langle e^{i\theta}\rangle_T\langle e^{-i\theta}\rangle_T]_c$ dominates at finite temperature) are unaffected by the applied field **H**. We, therefore, identify the line (11) with a transition from macroscopic superconductivity to spin-glass order. At finite temperature and fixed $H>0$, the surfaces $e_{1,0,\ldots,0}=0$ and $e_{1,-1,0,\ldots,0}=0$ determine respectively the transitions from the normal to the Abrikosov and spin-glass phases whereas the intersection of these two surfaces defines a line of multicritical points where the superconducting, spin-glass, and normal phases meet [Figs. 2(a) and 2(b)]. It is straightforward to verify that nonlinear terms in the expansion of H_r preserve the decoupling of the spin-glass fields $(S_{\mathbf{k}}=0)$ from those with $S_{\mathbf{k}}\neq 0$. That is to say, order in the Edwards-Anderson field induces order in all modes for which $S_{\mathbf{k}}=0$ but not those for which $S_{\mathbf{k}}\neq 0$. The interaction terms affect only the precise location of the phase boundary.

To obtain the detailed properties of these phases within mean-field theory, we have solved the Landau-Ginzburg equations resulting from saddle minimization with respect to both $\psi_{\mathbf{k}}$ and $\mathbf{A}^\alpha$ of Eq. (7). We restrict our attention to solutions which preserve replica symmetry, i.e., $\mathbf{A}^\alpha=\mathbf{A}$. In what follows, **A** and $\psi_{\mathbf{k}}$ are understood to represent equilibrium expectation values of these fields. We have identified three distinct types of solutions to these equations apart from the trivial one $\psi_{\mathbf{k}}=0$, $\nabla\times\mathbf{A}=\mu_0\mathbf{H}$ describing the normal phase:

(i) For sufficiently weak applied fields $(H<H_{c1})$, as will be clarified shortly, the energetically favorable solution is that for which $\mathbf{A}=0$ (Meissner effect in London gauge) and $\psi_{\mathbf{k}}(x)$ is spatially uniform and precisely that of a dilute XY ferromagnet. As in usual superconductors, this is accompanied by screening surface currents which decay on the scale of the London penetration depth λ, from the sample boundary. In the London gauge, the phase of $\psi_{\mathbf{k}}$ remains uniform and the supercurrent **J** is given by

$$\mathbf{J}=-\rho_s\frac{e^2}{mc}\mathbf{A};\quad \rho_s=C_2\lim_{n\to 0}\mu_0\frac{\tilde{T}}{n}\sum_{\mathbf{k}\neq 0}S_{\mathbf{k}}^2|\psi_{\mathbf{k}}|^2, \tag{12}$$

where $C_2=(da^d)^{-1}2ma^2/\hbar^2$ and ρ_s is the macroscopic superfluid density. The presence of the field **A** suppresses order in those modes for which $S_{\mathbf{k}}\neq 0$ so that at the glass transition ρ_s vanishes. In the weak-field limit $(A\to 0)$, the implicit dependence of $\psi_{\mathbf{k}}$ on **A** may be neglected and it may be shown[5] that the divergence of λ as $p\to p_c$ is governed by the conductivity exponent: $\lambda^{-2}(p-p_c)^t$. This equation implies that $\lambda^2/\xi_p^2\sim|p-p_c|^{(-t-2\nu)}$ is always large near threshold[12] for $d\geqslant 3$ ($t=3$ and $\nu=\frac{1}{2}$ in mean-field theory and $t=1.85$ and $\nu=0.85$ in $3d$). Thus sufficiently close to p_c, the granular medium behaves like a type-II superconductor. It follows that there is a critical field

$$H_{c1}=(\phi_0/4\pi\lambda^2)\ln(\lambda/\xi_p) \sim\mu_0(p-p_c)t \quad (T\to 0), \tag{13}$$

above which the Meissner state becomes unstable to the formation of an Abrikosov flux lattice (Fig. 1).

(ii) We have shown that the Landau-Ginzburg equations admit vortex solutions of the form

$$\psi_{\mathbf{k}}=\xi_p^{-2}f_{\mathbf{k}}(r)e^{imS_{\mathbf{k}}\theta}. \tag{14}$$

Here r and θ are cylindrical coordinates measured from the vortex core and m is a nonzero integer giving the number of flux quanta contained in the vortex. For small r, $f_{\mathbf{k}}(r)\sim r^{m|S_{\mathbf{k}}|}$ revealing that vortex cores retain spin-glass order at sufficiently low temperature.

(iii) For applied fields $H>H_g$, the energetically favorable solution to the Landau-Ginzburg equations is that in which complete flux penetration occurs

($\nabla x \mathbf{A} = \mu_0 \mathbf{H}$) and $\psi_{\mathbf{k}} = 0$ unless $S_{\mathbf{k}} = 0$. In the glass phase, we have shown that

$$\gamma_{ij}^{\alpha\beta}(q) = \delta_{ij} \lim_{n\to 0} \frac{\tilde{T}}{n} \sum_{S_{\mathbf{k}}=0} k^2 |\psi_{\mathbf{k}}|^2 = \delta_{ij}\gamma_g, \tag{15}$$

so that $Y_{ij} = 0$. Equation (8) implies that $\gamma_g \sim -[|\langle J(q)\rangle_T|^2]_c$ must be negative. Equation (15) does indeed predict a negative value for γ_g as can most easily be seen by considering the vicinity of the spin-glass to normal transition where only the $n(n-1)$ order parameters $\psi_{1,-1,0,\ldots,0}$ contribute to the sum. As in the normal and Meissner phases the net macroscopic supercurrent in equilibrium $[\langle J\rangle_T]_c$ is zero. However, unlike these more familiar phases, the spin-glass state is characterized by a randomly oriented distribution of frozen-in supercurrents as revealed by the nonzero value of γ_g. These frozen supercurrents lead to long-range fluctuations in the equilibrium magnetic field in the glass phase. Using $\delta\mathbf{B}(q) = i\mathbf{q}x\delta\mathbf{A}(q)$ and Eqs. (9) and (15), we obtain

$$\langle \delta\mathbf{B}^\alpha(q)\cdot\delta\mathbf{B}^\beta(-q)\rangle = [\langle|\delta\mathbf{B}(q)|^2\rangle_T]_c + (1-\delta_{\alpha\beta})[|\langle\delta\mathbf{B}(q)\rangle_T|^2]_c = 4\pi\mu_0 k_B T(\delta_{\alpha\beta} + 4\pi\mu_0|\gamma_g|q^{-2}). \tag{16}$$

Thus, in the glass phase, there is flux penetration with a uniform nonzero $[\langle B\rangle_T]_c$ everywhere but with strong local fluctuation in $\mathbf{B}$ leading to power-law ($x^{-(d-2)}$) decay in both $[\langle\delta\mathbf{B}(x)\cdot\delta\mathbf{B}(0)\rangle_T]_c$ and $[\langle\delta\mathbf{B}(x)\rangle_T\cdot\langle\delta\mathbf{B}(0)\rangle_T]_c$.

We mention finally that for the results described above, we have only considered solutions to the Landau-Ginzburg equations which do not break replica symmetry.[13] Although broken-symmetry solutions will not alter the qualitative features of the phase diagram discussed here, as in more traditional spin glasses they are important in distinguishing equilibrium from nonequilibrium properties of the glass phase.[14]

From an experimental point of view, samples of independent Josephson-coupled grains of the size of the zero-temperature superconducting coherence length are most likely to be produced in a controlled manner in two dimensions. It would be of interest, however, to study well-characterized three-dimensional composites where the predictions of our mean-field theory become more accurate. Since the supercnducting order parameter is experimentally inaccessible, we feel that the strongest signatures of the glass phase are likely to be in its dynamical properties. As in magnetic spin-glasses,[14] nonergodic behavior is likely to be manifest in differences between field-cooled and zero-field–cooled samples. If nonequilibrium metastable states are important, a conductivity experiment may determine whether the macroscopic superfluid density which we identified as being zero in the glass phase (assuming replica symmetry) exhibits remanence when this phase is entered from the Abrikosov phase. Dynamical properties such as ac conductivity may also probe the nature of barriers separating metastable states and the associated distribution of relaxation times.

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