Diamagnetic susceptibility of superconducting clusters: Spin-glass behavior

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We present a theory for the diamagnetic response of weakly linked superconducting clusters. In the model, superconducting grains, each small compared to a penetration depth, are weakly coupled into closed loops. These support screening supercurrents in response to an external magnetic field. In a magnetic field, a large cluster can support many supercurrent-carrying states of nearly equal energy, but energy barriers between these states tend to inhibit hops from one state to another at low temperatures. The picture is similar to that often proposed for spin glasses. An important consequence is predicted to be a large difference between the dc and ac susceptibilities at low temperatures. The former, an equilibrium property, will fall off much more rapidly with field than the latter, which is generally a property of the metastable states. In addition, the magnetization of a cluster varies discontinuously with field; for a sufficiently large cluster, the magnetization is everywhere discontinuous. To check these conjectures, two examples are studied. The first consists of single loops of random areas and orientation, which can be solved analytically at zero temperature. The second involves random two-dimensional clusters of many closed loops, and is studied via careful Monte Carlo simulation at various temperatures and fields. Both examples display the expected strong differences between ac and dc susceptibilities at low temperatures. Our predictions are found to be quite similar to the experimental results of Bastuscheck *et al.* [Phys. Rev. B **24**, 6707 (1981)] for the fibrous superconductors $NbSe_3$ and $TaSe_3$.

I. INTRODUCTION

Bulk superconductors respond characteristically to an external magnetic field. Type-I superconductors totally exclude magnetic flux (except for a surface layer of thickness of order the penetration depth) below the transition temperature and up to a critical field H_C. Type-II superconductors totally exclude flux up to a field H_{c1} and partially until the upper critical field H_{c2}. In contrast to such bulk materials, the behavior of composite superconductors is much more complex. A variety of experiments have indicated unusual behavior of the magnetic susceptibility, the upper critical field, field-dependent magnetization, and penetration depth in such materials.[1–7]

This paper proposes a theory of the diamagnetic response of certain superconducting composites. The materials we consider consist of isolated clusters of superconducting grains, each smaller than the penetration depth, embedded in a nonsuperconducting host, and weakly coupled together by the proximity effect (if the host is a normal metal) or Josephson tunneling (for an insulating host). Materials of this kind can readily be prepared in the form of two-dimensional films by modern photomicrolithographic techniques. The geometry can be designed almost to specification and the host can be either a normal metal or an insulator.[8–11] Our model is probably also reasonable for many three-dimensional composites. Often, of course, the particles in a composite are actually coupled into an infinite cluster rather than many finite clusters. We have considered finite clusters in part for computational convenience. Many of the properties of these finite clusters, however, should also be observable in infinite clusters.

The magnetic properties of random composite superconductors have already been the subject of several theories.[12–19] Rammal *et al.*,[13,14] Straley and Visscher,[17] de Gennes,[12] Alexander,[19] and Stephen,[18] among others, consider the properties of a network of superconducting wires, each thin compared to a penetration depth, in an applied field. They typically use scaling arguments to predict low-field susceptibilities near the percolation threshold p_c. This is the concentration at which an infinite connected path of wires first forms. They define a new critical exponent, for the divergence of the diamagnetic susceptibility near p_c and relate it to other exponents. This same exponent can also be calculated for clusters of weakly connected grains.[15] Shih *et al.*[16] consider the phase diagram of such random materials in a magnetic field, and find many analogies with a spin glass. Here, we extend this paper to treat diamagnetic susceptibility. Unlike most previous papers, we concentrate on strong-field behavior where the spin-glass features are most obvious.

The remainder of the paper is organized as follows. Section II describes our model, and qualitatively analyzes its diamagnetic properties. Various spin-glass features are predicted. These include differences between dc and ac magnetic susceptibility, and between magnetic behavior resulting from cooling in a field and cooling in zero field. Section III applies the model to two types of clusters. The first is a ring of weakly coupled grains (for which the susceptibility can be found analytically at temperature $T=0$). The second type is a random two-dimensional cluster. Its behavior is complicated and must be obtained numerically, though it resembles that of a ring. Finally, Sec. IV gives a discussion of the results and of their connection to experiment.

II. THE MODEL AND ITS QUALITATIVE BEHAVIOR

The clusters of interest contain N superconducting grains embedded in a nonsuperconducting host. The ith grain is centered at $\vec{x}_i$ and has a complex energy gap $\psi_i = \Delta_i \exp(i\phi_i)$. The grains are weakly coupled via the host according to the Hamiltonian[20–22,16]

$$\mathscr{H} = -\sum_{<ij>} J_{ij}\cos(\phi_i - \phi_j - A_{ij}) . \quad (1)$$

Here J_{ij} is the coupling energy between grains i and j, and the phase factor A_{ij} is given by

$$A_{ij} = \frac{2\pi}{\Phi_0}\int_i^j \vec{A}\cdot d\vec{l} . \quad (2)$$

In Eq. (2), $\Phi_0 = hc/2e$ is an elementary flux quantum, and the line integral is taken along a line joining the centers of grains i and j.

The coupling energy appearing in Eq. (1) is

$$J_{ij} = \frac{h}{2e} I_{ij} , \quad (3)$$

where I_{ij} is the superconducting critical current between grains i and j. For a Josephson junction between two identical grains, this current is[23]

$$I_{ij} = \frac{\pi}{2e}\frac{\Delta(T)}{R_{ij}}\tanh\left[\frac{\Delta(T)}{2k_BT}\right] , \quad (4)$$

where R_{ij} is the resistance between grains i and j in their normal state and T is the absolute temperature. If the coupling is induced by the proximity effect through a normal metal, Eq. (4) is replaced by[10]

$$I_{ij} = C(1 - T/T_{cs})^2 \exp[-r_{ij}/\xi_n(T)] , \quad (5)$$

where C is a constant, T_{cs} is the transition temperature of the superconducting grains, r_{ij} is the separation between grain centers, and $\xi_n(T)$ is the coherence length of the normal metal. In this paper, for clarity, we shall neglect the temperature dependence of the coupling. Thus we emphasize effects due to the weak coupling between the grains.

The thermodynamic properties of this model are obtained by treating the phases as classical variables within the canonical ensemble. Hence the Helmholz free energy is given by

$$F = -k_BT\ln Z , \quad (6)$$

$$Z = \int \left[\prod_i d\phi_i\right] \exp(-\mathscr{H}/k_BT) . \quad (7)$$

In terms of F, the magnetic moment μ is

$$\mu = -\left[\frac{\partial F}{\partial H}\right]_T , \quad (8)$$

where H is the external magnetic field. Similarly the isothermal differential susceptibility is

$$\chi_{\rm ac} = \left[\frac{\partial M}{\partial H}\right]_T , \quad (9)$$

where M is the magnetic moment per unit volume.

Both of the derivatives (8) and (9) can be computed numerically from the free energy. It is easier, however, to find the magnetic moment for each cluster directly from the circulating supercurrents. Thus the moment μ of a cluster is

$$\vec{\mu} = \frac{1}{2c}\sum_{<ij>} \langle \vec{X}_{ij} \times I_{ij}\vec{x}_{ij} \rangle , \quad (10)$$

where

$$I_{ij} = \frac{2eJ_{ij}}{h}\sin(\phi_i - \phi_j - A_{ij})$$

is the Josephson current from grain i to grain j, $\vec{X}_{ij} = (\vec{x}_i + \vec{x}_j)/2$ is the vector joining the origin to the position of the midpoint between grains i and j, and $\vec{x}_{ij} = \vec{x}_j - \vec{x}_i$ is the vector distance from grain i to grain j. The magnetic moment of a finite cluster, of course, does not depend on the choice of origin in this expression, and so Eq. (10) is completely unambiguous. The canonical average of an operator $\theta(\phi_1, \ldots, \phi_n\rangle$, denoted $\langle\theta\rangle$, such as that given in Eq. (10), is computed from the relation

$$\langle\theta\rangle = Z^{-1}\int \theta(\phi_1, \ldots, \phi_N)\left[\prod_i d\phi_i\right]\exp(-\mathscr{H}/k_BT) . \quad (11)$$

The key to this model is "frustration."[24] By frustration, we mean the fact that at finite fields, any cluster with closed loops cannot find a state which simultaneously minimizes all the bond energies. The frustration is produced by the phase factors A_{ij}, which make some of the bonds ferromagnetic (i.e., favoring equal phases ϕ_i and ϕ_j), some antiferromagnetic, and most favoring an angle between these extremes. As in more familiar frustrated systems (e.g., magnetic models), a large frustrated cluster, with many closed loops, can choose among numerous competing ground states with nearly equal energy. In a finite cluster, only one of these is the true ground state. But others will lie only a small energy above it. As the field is varied, the various levels will cross one another. The cluster must hop from one configuration to another in order to stay in the ground state. Each such hop will be accompanied by an abrupt change in magnetization. In a large cluster, these flux hops will be very closely spaced in field. The curve of ground-state energy $E(H)$ will consist of many short arcs, with discontinuities of slope where they join.

This picture applies only to a cluster in equilibrium. But at low temperatures, the cluster will probably be trapped in a metastable configuration and the measured properties will be metastable ones. In order to reach its ground state, the cluster must climb an energy barrier which is too high to overcome at low temperatures. The cluster can escape and equilibrate, however, if it is sufficiently heated.

This picture leads to a difference between the dc and ac diamagnetic susceptibilities. The former is defined by the relation

$$\chi_{\rm dc} = +(M/H)_T , \quad (12)$$

and is a true equilibrium property. χ_{ac}, defined in Eq. (9), can be measured by superimposing on a fixed dc field a small-amplitude parallel ac field. If the frequency is larger than the relaxation rate of the cluster, χ_{ac} measures a nonequilibrium property. It is also typically larger than χ_{dc}. These differences are also characteristic of more usual spin glasses.[25] A real composite will have a broad distribution of relaxation rates. At high frequencies, more of the clusters exhibit a nonequilibrium response to an external field than at low frequencies. The ac susceptibility should therefore increase (in absolute value) with increasing frequency. At higher temperatures, trapping into metastable states should be less important and this frequency dependence should be weaker.

If the clusters are cooled slowly in a fixed field, the resulting configuration should be in equilibrium, because the slow cooling allows the cluster to be annealed at relatively high temperatures. Cooling in zero field, and turning on the field at fixed low temperature, may result in a metastable magnetization, since the cluster has no time to equilibrate as the field is turned on. This distinction between field cooling and zero-field cooling has often been noted in bulk spin glasses.[26] Similarly, if the clusters are field-cooled to a low temperature and the field is suddenly turned off, there will remain trapped flux which slowly decays away as the cluster equilibrates. This trapped flux is the superconducting analog of remanence in magnetic systems. Its decay should have an anomalous (i.e., nonexponential) time dependence characteristic of a very broad distribution of relaxation times, just as is seen in true spin glasses.[27]

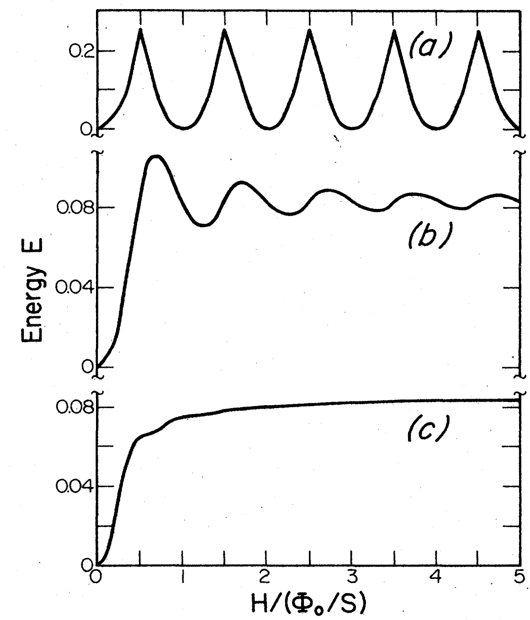

FIG. 1. Internal energy E per loop at temperature $T=0$ versus external magnetic field H, for (a) an assembly of superconducting loops of projected area S perpendicular to the field; (b) an assembly of loops of area S randomly oriented relative to the field. In (c) the loops are randomly selected to have one of 20 different areas equally spaced from $0.1S$ to $2S$. The energy is given in units of $2J\pi^2/N$ where J is the coupling constant and N is the number of grains in the loop. $\Phi_0=hc/2e$ is a flux quantum. Note differences in vertical scales.

III. NUMERICAL EXAMPLES

A. Single loops

All of these features are easily seen in a simple example of a granular system which can be solved analytically—randomly oriented loops of grains in a magnetic field. Consider a loop of N identical superconducting grains, with identical coupling J between nearest neighbors, and assume initially that they are oriented perpendicular to the applied magnetic field B. In the most convenient gauge, the phase factors A_{ij} are given by

$$A_{ij}=\frac{2\pi}{N}\frac{\Phi}{\Phi_0}\,, \tag{13}$$

where Φ is the flux through the loop. At $T=0$, current conservation requires that all the phase differences $\phi_{i+1}-\phi_i$ be equal. Continuity requires that they sum to an integral multiple of 2π. The loop must choose among various metastable current-carrying states that satisfy these two conditions. The stable one at any field is that with the lowest energy. The ground-state energy $E(H)$ obtained in this way is plotted in Fig. 1(a). In making this plot, we have assumed that the number of grains N in a loop is large, so that the cosine factors in the energy can be expanded as $\cos(\phi_{ij}+A_{ij})=1-\frac{1}{2}(\phi_{ij}+A_{ij})^2$. The curve of Fig. 1(a) thus consists of arcs of parabolas.

In Fig. 1(b) we show $E(H)$ for a distribution of loops, each of same area but oriented at random, so that the projected area perpendicular to the field varies. The discontinuities observed for single loops are washed out, but the energy still displays oscillations, the vestiges of the slope discontinuities of Fig. 1(a). Figure 1(c) shows the effects of averaging over loops of different areas as well as orien-

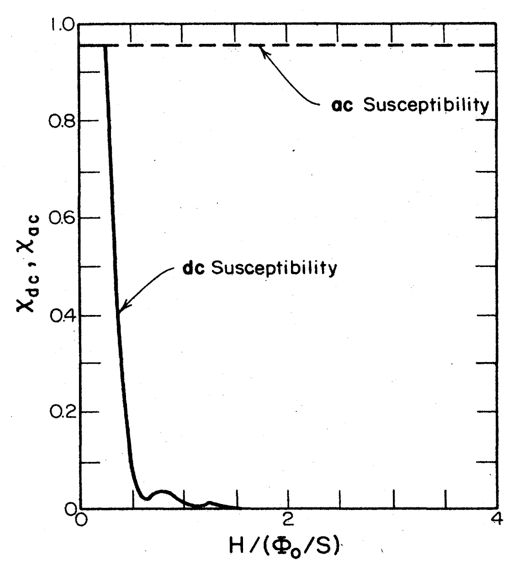

FIG. 2. Direct-current and alternating-current susceptibilities for the loops of Fig. 1(c). Susceptibilities are given in units of $2nJ\pi^2/[N/(\Phi_0/S)^2]$, where n is the number of loops per unit volume and the other symbols are as in Fig. 1.

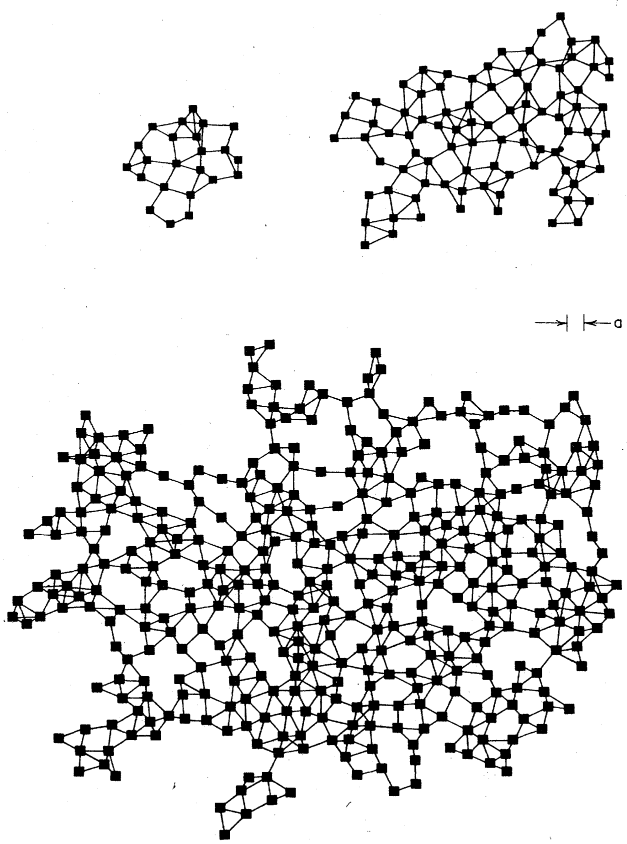

FIG. 3. Random two-dimensional clusters containing 23, 89, and 385 grains, used in the calculations shown in Figs. 4—6. The length a is the grain diameter.

tations. Now the oscillations have disappeared and $E(H)$ rather smoothly approaches a limiting value at high field.

Figure 2 shows the dc and ac susceptibilities for the loops of Fig. 1(c). The ac susceptibility is essentially an average over area and orientation of the second derivative of the curve in Fig. 1(a). Since this is a constant, the ac susceptibility is strictly independent of field. In contrast, the dc susceptibility [Eq. (12)] is strongly field-dependent and falls rapidly to zero with increasing field. Thus this simple analytic model already displays the most prominent features of the more complex loops discussed below.

B. Random clusters

The same behavior is displayed by more complicated finite clusters of superconducting grains typical of a real composite. We have generated the clusters by a computer algorithm which preserves the positional disorder of real composites below percolation. The method is to add points to a square "substrate" of edge L. Each successive point is deposited at a random position, but is rejected if it falls within a distance a of a previous point. This length a is thus the grain diameter. The interactions are chosen equal to J if the grain centers lie within $2a$ of each other, zero otherwise. To prevent edge effects, periodic boundary conditions are assumed. For most calculations, points are added until a number of large but finite clusters are generated. Since only closed loops of grains contribute to the magnetic moment, dangling ends of each cluster are stripped off before their properties are computed.

The actual evaluation of the canonical averages at finite T is carried out using standard Monte Carlo techniques within the Metropolis algorithm. The numerical effect of frustration is that in the simulations (as in experiment) the clusters tend to sit in metastable configurations. It is therefore necessary to cycle the temperature up and down in various ways, annealing at high temperatures and slowly cooling, in order to be reasonably sure that the resulting averages really refer to equilibrium states. Since the results we quote are reproducible, we have confidence that they do indeed correspond to equilibrium.

Figure 3 shows several typical clusters generated by the method just described, minus dead ends; the quoted "cluster" number N includes only those grains in the looped

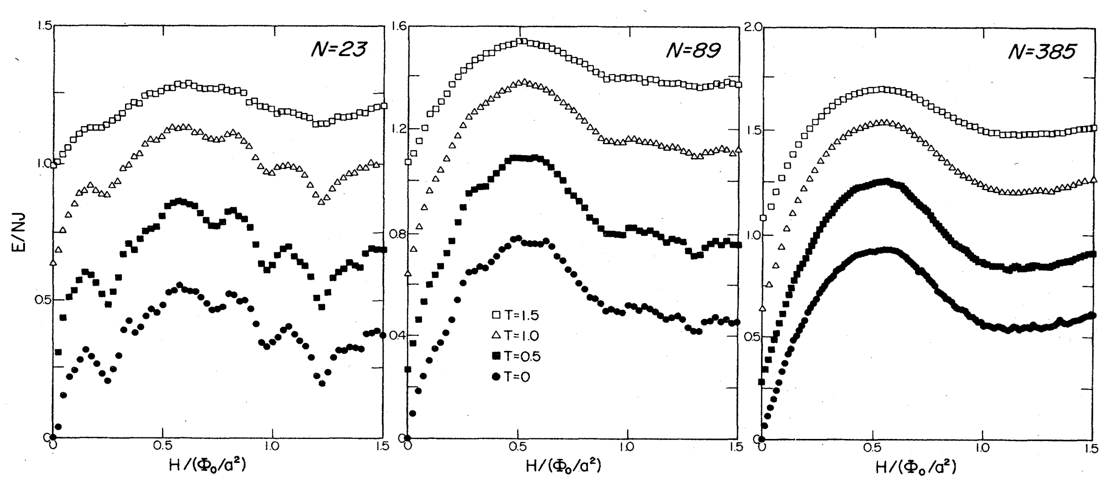

FIG. 4. Internal energy per grain as a function of field for the clusters of Fig. 3, calculated by Monte Carlo methods at several different temperatures. The temperatures T are measured in units of J.

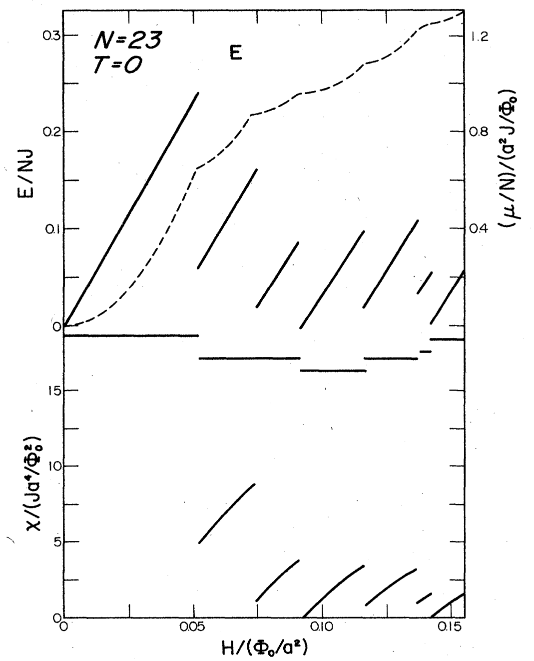

FIG. 5. Upper half of figure: Energy E at $T=0$ (dashed line) and magnetic moment (solid diagonal line segments) as a function of magnetic field H for the 23-grain cluster. Lower half of figure: ac susceptibility (horizontal line segments) and dc susceptibility (diagonal solid segments) for the $N=23$ cluster. The first horizontal segment represents both the ac and dc susceptibilities for those fields.

portion of a cluster. Each cluster typically has several loops of different areas, most of order a^2 or larger.

The internal energy of the clusters of Fig. 3 is plotted versus field at several temperatures in Fig. 4. At $T=0$, the energy is actually only a piecewise differentiable function of magnetic field. This is clearest for the smallest cluster shown (N=23) but is true for the larger ones also. The continuous regions extend over a larger range of fields for smaller clusters, and their discontinuities in slope are also larger. At $T>0$ the observed energy is simply a thermal average of all available energy states weighted by the appropriate Boltzmann factor. The discontinuities in slopes are hence washed out and the energy varies smoothly with field.

The discontinuities in slope are made even more apparent in Fig. 5, which displays the magnetization $M(H)$ for the $N=23$ cluster at $T=0$, along with χ_{dc} and χ_{ac}. $M(H)$ resembles that of single loops of Fig. 1(c). χ_{ac} is nearly field independent for the fields considered, as in the single-loop case, while χ_{dc} falls off very rapidly with field (as well as exhibiting the same nonanalyticities as the magnetization itself).

The magnetization $M(H,T)$ is shown for all three clusters in Fig. 6. The curves are the results of an explicit Monte Carlo calculation of the magnetic moment [Eq. (10)]. At $T=0$, but not at finite temperature, the magnetization could also be obtained by numerical differentiation of the energy.

At finite temperatures, all the energy curves, for all the clusters we have examined, display very much the same shape except at very low fields. They rise quadratically with field, approach a maximum at a field of order $\frac{1}{2}$ flux quantum per a^2 of area, then fall off slightly and saturate. The same is true of the randomly oriented loops of Sec. III A. At low temperatures, there are also the discontinuities of slope mentioned above. The quadratic dependence of energy on field at low fields is just the result of a constant diamagnetic susceptibility $(\partial M/\partial H)_T$. At such low fields the dc and ac susceptibilities of these clusters become equal. The strong-field saturation of energy is due to "saturation of frustration:" At high fields, the phase factors A_{ij} are randomly distributed over an interval of width at least 2π and further increases in field do not change this distribution substantially. The internal energy therefore approaches a constant value. If the clus-

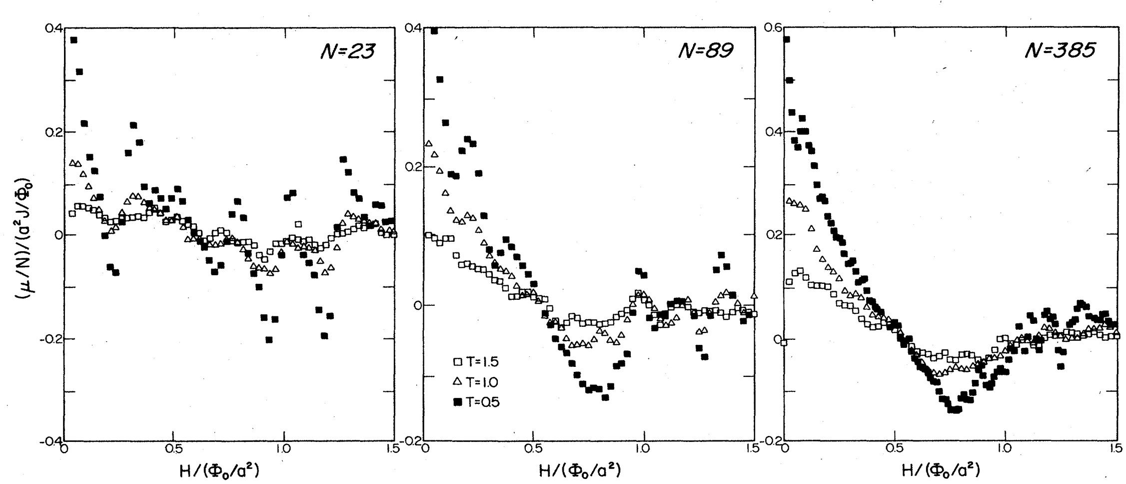

FIG. 6. Magnetic moment per grain as a function of field for the clusters and temperatures shown in Fig. 4, as calculated by Monte Carlo simulation.

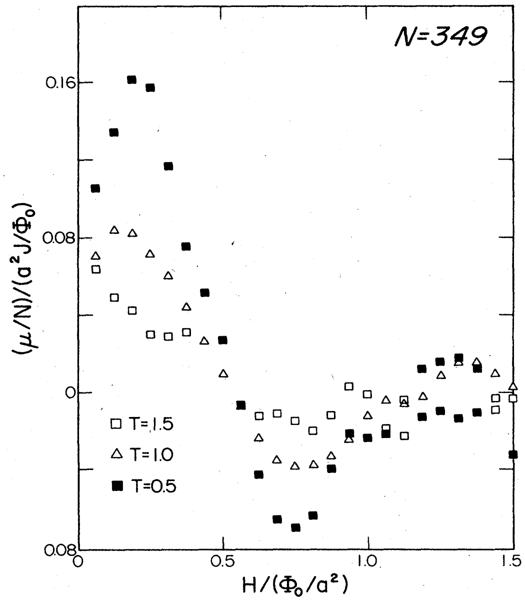

FIG. 7. Magnetic moment per grain for a two-dimensional "amorphous" sample below percolation, as calculated by Monte Carlo simulation at several temperatures. The areal fraction occupied by the grains is 0.171.

ter consisted of a single loop, the internal energy would vary periodically with field, with a period of one flux quantum per area of the loop. Since the cluster actually consists of several loops with mutually incommensurate areas, the real behavior of the energy is a superposition of this periodic behavior with a number of different periods.

The universal behavior of the energy is mirrored in the magnetization which at any temperature rises initially linearly with field to a maximum value which is about the same for clusters of any size, then falls off towards zero at higher fields. At the stronger fields, the magnetic moment usually exhibits damped oscillations. The initial slope is larger, and the field for which the maximum in the magnetization occurs is smaller, for the larger clusters, because larger clusters have a larger low-field susceptibility than do smaller ones.

A real two-dimensional sample below the percolation threshold will, of course, consist of a variety of finite clusters of various sizes. If the dipolar coupling between the diamagnetic clusters can be neglected, the total magnetic moment in this case will simply be a sum of the magnetic moments of the various individual clusters. This averaging over clusters will further wash out the discontinuities in magnetization, even at low temperatures. The resulting variation of magnetization with field is shown in Fig. 7 for several different temperatures in a typical two-dimensional sample of disk-shaped grains computer generated according to the prescription given earlier. The particular sample shown corresponds to a filling fraction (i.e., areal fraction occupied by the disks) of 0.171. The universal behavior mentioned earlier is even more clearly apparent than for the individual clusters shown in the earlier figures.

IV. DISCUSSION

To compare the present results with experiment, we must express them in experimental units. Consider first the isolated loops discussed in Sec. III A. If there are n such loops per unit volume, each of projected area S perpendicular to the field, then the low-field diamagnetic susceptibility in the absence of screening effects is

$$-\chi_{\mathrm{ac}} = \frac{J}{\Phi_0^2} n S^2 , \tag{14}$$

where Φ_0 is a flux quantum. The field at which the first flux slip occurs, that is, where the ac and dc susceptibilities start to differ, is

$$H_{c1} = \Phi_0 / 2S . \tag{15}$$

For loops of area $100\mu^2$, $H_{c1} \sim 0.1$ g. If the coupling energy $J \sim 10$ K, then the zero-field susceptibility will approach unity when $n \sim 10^7$. At such densities, local-field corrections must be applied to the susceptibility.

The results for large clusters (Sec III B) are quite similar to those of single loops of increasing area. The zero-field susceptibility is larger for the larger clusters (increasing roughly as the radius of the clusters). This is consistent with the result that the susceptibility of a two-dimensional composite diverges as the percolation threshold is approached from below (i.e., as the average cluster size approaches infinity). Similar results are expected for three-dimensional clusters, possibly corrected for screening of the applied magnetic field in sufficiently large clusters (because of a finite penetration depth).

The experiments most relevant to the present results are those reported by Buhrman and collaborators on the fibrous superconductors $NbSe_3$ and $TaSe_3$. These materials are bundles of superconducting fibers, each thin compared to a penetration depth and embedded in a nonsuperconducting (probably a dielectric) host. They remain resistive even at very low temperatures, indicating that the superconducting elements are coupled only as finite clusters. The dc susceptibility is generally much smaller than the ac susceptibility and falls off much more rapidly with field, as is predicted by the nonequilibrium "spin-glass" model given here. The field characterizing the falloff appears to be about 0.1 g, corresponding to loops of area 100 μ^2, a not unreasonable value for these samples. Flux trapping upon turning off the field at low temperatures is also observed, indicating remanence in a metastable state. The way this trapped flux decays with time would give information about the distribution of relaxation times in this system. The magnitudes of the observed susceptibilities, which vary from 0.001 to the theoretical maximum of $(1/4\pi)$, seem consistent with the likely density of clusters in the samples.

Although our results here are for finite clusters, they are probably applicable to infinite clusters also, i.e., to bulk systems. For very large clusters and for bulk samples, the discontinuities in magnetization become so close as to form an apparently continuous curve, but still with a large distinction between ac and dc properties. Thus we expect the same kind of diamagnetic response in an infinite composite (in which the resistivity approaches zero at low temperatures) as in one comprised of finite clusters.

It is of some interest to estimate the relaxation rates involved in the decay of metastable states. A reasonable guess would be

$$W = W_0 \exp(-\Delta E / k_B T) , \tag{16}$$

where W_0 is an attempt frequency and ΔE is an energy barrier. Typical energy barriers would be of the order of the intergrain coupling energy J, but they would almost certainly have an enormous variation, down to very minute values. For tunnel junctions a reasonable guess for the attempt frequency is $W_0 \sim eI_c R/\hbar$ where I_c is the critical current of the tunnel junction and R is the normal-state resistance. For typical composites $W_0 \sim 10^m$ with $m \sim 10-12$. Thus we expect a broad spectrum of relaxation rates as high as 100 Ghz or so, but decreasing to far below 1 Hz at low temperatures.

To summarize, we have reported in this paper a model for the diamagnetic properties of a composite superconductor. The composite consists of isolated clusters of superconducting grains coupled together by Josephson tunneling or the proximity effect. Calculations indicate that such materials will have diamagnetic properties similar to those observed in other glassy systems. These include strong differences between dc and ac susceptibilities, remanent magnetization in zero field with anomalous time dependence, and a variation of energy with magnetic field similar to a system undergoing a nearly continuous series of weakly first-order phase transitions. The predictions are consistent with the limited available experimental data, and are relevant to many other experiments that could be readily carried out.

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