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Metastable effects and random polarities of magnetic moments in disordered systems of Josephson junctions

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We use the Monte Carlo technique to simulate experiments on zero-field-cooled (ZFC) and field-cooled (FC) superconducting samples. As our model we take a three-dimensional, strongly disordered system of Josephson junctions. For fields exceeding a certain critical value H_{c1}, we find well-pronounced differences between ZFC and FC magnetizations. At low temperatures, the diamagnetic response of FC samples cooled at H_{c1} is reduced due to the polarity changes of many local magnetic moments.

There has been a growing interest in both experimental[1-4] and theoretical[4-9] investigations of systems of superconducting grains coupled via Josephson junctions. It has been argued that at low temperatures, due to the frustration induced by an external magnetic field, such systems may undergo a transition into the superconducting-glass state.[8] More recently, Bednorz, Takashige, and Müller[10] suggested that irreversible effects observed in high-T_c superconductors may also occur due to the formation of the superconducting-glass state, i.e., superconducting clusters coupled by Josephson junctions may be formed inside a sample. For single-crystal samples, Yeshurun and Malozemoff[11] proposed an alternative explanation in terms of the "giant-flux-creep" mechanism. Nevertheless, the existence of Josephson junctions in superconducting ceramics has been experimentally confirmed.[12,13]

One of the characteristic metastable effects observed in high-T_c superconducting materials is the existence of differences between magnetic properties of the zero-field-cooled (ZFC) and field-cooled (FC) samples.[10,11,14] This effect was qualitatively reproduced in a recent Monte Carlo (MC) simulation[15] of a weakly disordered system of Josephson junctions. The applicability of the superconducting-glass model to high-T_c superconductors was criticized in Ref. 14. There it was argued that due to the random orientations of local currents the resulting local magnetic moments in FC samples may have random polarities. It means that a large fraction of local magnetic moments may be paramagnetically oriented. Thus, contrary to experiment, there should be no correlation between the remnant magnetization ($M_{\rm rem}$) and the values of ZFC and FC moments. The above conclusions were drawn from the analysis of a simple model consisting of a single ring of superconducting grains.

In this paper we use MC simulations to investigate a more realistic, three-dimensional, strongly disordered systems of Josephson junctions. We concentrate on metastable effects such as differences in magnetic response of ZFC and FC samples. We find that strong disorder does not destroy qualitative features discussed in Ref. 15. We also confirm observations made in Ref. 14, i.e., we find that indeed in certain magnetic fields local magnetic moments in FC samples are oriented paramagnetically. However, our preliminary analysis of the role of the system size indicates that this effect may disappear in the thermodynamic limit.

As a model we take a system[16] of N superconducting grains embedded in a nonsuperconducting host. Grains are distributed over ten evenly spaced planes parallel to the x-y plane. For each grain the (x,y) coordinates and a plane number are chosen randomly. The grain is added to the system when its distance to all other grains in the same plane is bigger than a_r and is discarded otherwise. As a consequence, the resulting structure is characterized by a strong, uncorrelated positional disorder.[17] We assume that below T_{cs}—the temperature of superconducting transition of a single grain—tunneling junctions are formed between grains in the same plane if their distance is less than $2a_r$. Grains from two adjacent planes are coupled if their projections on the x-y plane are not further apart than $2a_p$. The same rules are used in imposing periodic boundary conditions. We choose $a_p=0.3a_r$, and the distance between planes is taken equal to a_r. All "loose ends," i.e., strings of coupled grains which do not form closed loops, are removed. Finally, we check that the generated structure cannot be decomposed into independent substructures without cutting at least one link.

The system of superconducting grains coupled via tunneling junctions is described by the Hamiltonian[7,18]

$$\mathcal{H}=-\sum_{\langle i,j\rangle} J_{ij}\cos(\phi_i-\phi_j-A_{ij})\,, \qquad (1)$$

with

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

where $\mathbf{A}$ is the magnetic vector potential, Φ_0 is the magnetic flux quantum, and ϕ_i and ϕ_j denote phases of the superconducting order parameter within the ith and jth grains. The integral in Eq. (2) is taken along a straight line and the sum runs over all coupled pairs of grains. As a simplification we assume that all $J_{ij}=J$ and that the value of J does not depend on temperature and magnetic field. We further take $\mathbf{A}$ as the vector potential of the homogeneous field, that is we neglect local-field changes due to the superconducting currents between grains.

In order to study thermodynamic properties of the system we consider phases ϕ_i as classical variables and use the standard Metropolis algorithm to compute thermal averages $\langle\ \rangle$ within a canonical ensemble. We are mainly interested in the value of the magnetic moment per grain, which is defined as a thermal average of

$$\mathbf{M}=\frac{\pi J}{2\Phi_0 N}\sum_{\langle ij\rangle}\sin(\phi_i-\phi_j-A_{ij})(\mathbf{x}_i-\mathbf{x}_j)\times(\mathbf{x}_i+\mathbf{x}_j)\,, \quad (3)$$

where $\mathbf{x}_i$ is a position of the grain i. In addition, we also compute the specific heat defined as

$$c_H=\frac{1}{NkT^2}(\langle\mathcal{H}^2\rangle-\langle\mathcal{H}\rangle^2)\,. \quad (4)$$

To simulate the ZFC and FC behaviors, we follow the procedure used in actual measurements. First, we turn off the magnetic field and cool the system from $T=4$ to $T=0.01$ (in units J/k, where k is the Boltzmann constant). Then we slowly turn on the magnetic field and equilibrate our system keeping $T=0.01$. Next, we fix the field value and slowly increase temperature from $T=0.01$ to 3.5. This part of our calculation corresponds to a ZFC experiment. Subsequently, without changing the field, we reverse the process and decrease temperature to its initial value 0.01, simulating an FC experiment. At each temperature we perform 60000 Monte Carlo steps (MCS) and use last 40000 MCS to compute thermal averages. In one MCS all phases are updated once and after the initial equilibration every second MCS is used in computing thermal averages.

To understand qualitatively the behavior of our system we first perform simulations of ZFC-FC properties of a single cluster ($N=381$) in various magnetic fields, $H=0.01,0.02,\ldots,0.05$ in units of Φ_0/a_r^2. For $H=0.01$ we find that ZFC and FC properties are exactly equal. In all fields $H>0.02$ we find differences between ZFC and FC magnetizations similar to those observed in Ref. 15. For $H=0.02$, however, ZFC and FC magnetic moments differ in sign. This field corresponds to the strongest diamagnetic response of the ZFC sample at $T=0.01$ (such a value of the magnetic field we further denote by H_{c1}). In order to obtain statistically reliable results we repeat simulations for seven or five independent structures of the same size, choosing fields $H=0.03$ and 0.02, respectively. Averaged results are presented in Figs. 1 and 2.

Figure 1 shows the low-temperature behavior of the magnetic moment parallel to the external field computed for $H=0.03$. In both ZFC and FC cases our system responds diamagnetically, in accordance with expectations, with the ZFC magnetic moment being significantly smaller than the FC one for $T\leq 0.2$. Big error bars are

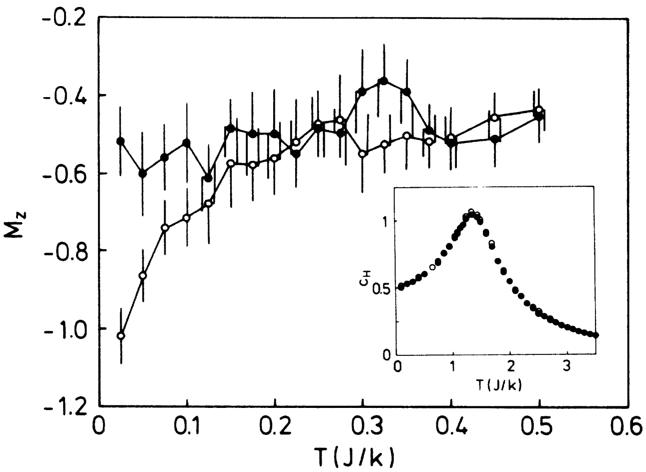

FIG. 1. Magnetic moment parallel to the field (in units of $\pi Ja_r^2/N\Phi_0$) as a function of temperature for ZFC (open circles) and FC (solid circles) experiments at $H=0.03\Phi_0/a_r^2$. Results are averaged over seven independent structures with $N=375$, 381, 384, 385, 381, 387, 383. The inset shows the corresponding specific heats computed in the whole temperature range. When not shown, the error bars are of the same size (or smaller) as the symbols used.

due to the fact that for $H>H_{c1}$ the amount of states with the same energy and different magnetic moments dramatically increases in each of the investigated systems. Corresponding results for $H=0.02$ are presented in Fig. 2. The ZFC-FC difference is here even more pronounced. Note that the positive, low-temperature, FC magnetic moment persists after averaging over independent structures. This behavior resembles results of the analysis presented in Ref. 14. Small error bars indicate the remarkable stability of the effect. For both fields the magnetic moment vanishes above some critical temperature T_c which coincides with the maximum of the specific heat (specific heat values are displayed as insets in Figs. 1 and 2).

To get an idea of how our results depend on the size of

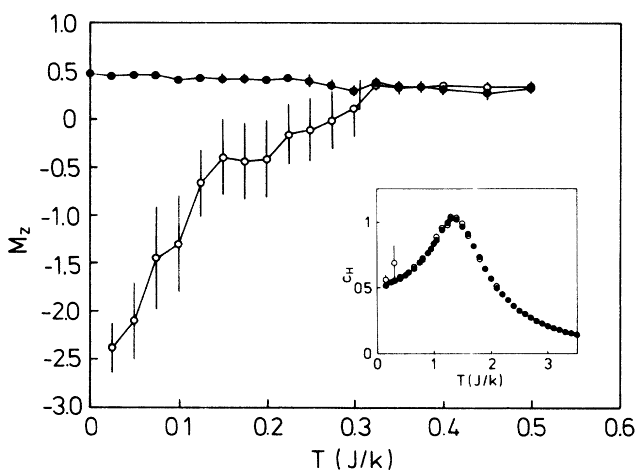

FIG. 2. The same as in Fig. 1 but for five structures with $N=381$, 384, 385, 381, 387 and $H=0.02\Phi_0/a_r^2$. Below this field irreversible effects disappear for samples with $N\simeq 400$.

our sample, we repeat similar simulations for a system consisting of $N=1163$ grains, in fields $H=0.01$, 0.02, and 0.03 (since the amount of computer time needed increases at least linearly with the number of links, we are not able to perform an ensemble average). In all cases we find well pronounced ZFC-FC differences but now the observed magnetic moments stay always negative. The minimal ZFC magnetization at $T=0.01$ occurs for $H=0.01$. Figure 3 shows the resulting magnetization in this field. The magnetic moment vanishes above $T_c \simeq 1.7$ which, as previously, coincides with the maximum of the specific heat. It is interesting to note the large jump of the FC magnetization around $T=0.5$. Below this temperature the diamagnetic response is substantially reduced, which means that with decreasing T many local currents change their polarities. These results are thus locally similar to the results we obtain for smaller structures and $H=0.02$. In both cases magnetic field is close to its critical value H_{c1}, above which the absolute value of magnetization (at $T=0.01$) starts to decrease. Below this field the behavior of our system is fully reversible, i.e., we observe no difference between ZFC and FC "experiments." The value of H_{c1} decreases with increasing N from $H_{c1} \simeq 0.02$ for $N \simeq 400$ to $H_{c1} \simeq 0.01$ for $N=1163$. For a system with $N \simeq 400$ it also decreases from $H_{c1} \simeq 0.03$ (see Ref. 16) to $H_{c1} \simeq 0.02$ when free boundary conditions are replaced with the periodic ones. This indicates that most probably $H_{c1} \rightarrow 0$ with $N \rightarrow \infty$. In the FC samples close to H_{c1} we observe that at low temperatures a great part of local magnetic moments becomes paramagnetically oriented (cf. Figs. 2 and 3). As a consequence, the overall FC response of smaller structures becomes paramagnetic or, as we see for the $N=1163$ sample, the diamagnetic response is strongly reduced. Observed properties of our systems strongly depend on the path of the process in the $H-T$ plane but we see practically no change when the steps in the values of T and H are reduced by a factor of 2.

Random polarity reversal of local magnetic moments was discussed by Malozemoff *et al.* in Ref. 14. They argued that this effect should result in a difference between the remanent magnetization of the sample ($M_{\rm rem}$) and the value of $M_{\rm FC}-M_{\rm ZFC}$. The observed absence of such a difference in actual measurements on single crystals and ceramics was then used as an argument against the application of the superconducting-glass model to the description of high-T_c superconductors. We note, however, that to realistically describe remanent magnetization it is necessary to take into account magnetic fields produced by tunneling currents. Otherwise, with the external field switched off, magnetic moments decay very fast to zero. But, as discussed earlier, we indeed see polarity reversal of local magnetic moments in our simulations. Finite-size scaling arguments quoted above suggest, however, that with increasing N the reversed moments make a vanishing contribution to the FC magnetization of the system. This means that measurements of the total magnetization are probably not sufficient to decide between two mechanisms proposed to explain irreversible effects in granular (ceramic) samples. On the other hand, as pointed out in Ref. 14, distribution of magnetic moments in the superconducting-glass picture has no necessary relation to the Abrikosov lattice of flux lines, since the characteristic length scale is defined here by the sizes of superconducting grains. Measurements of local magnetization should thus allow us to conclude to what extent the model discussed may be used to describe high-T_c ceramic superconductors.

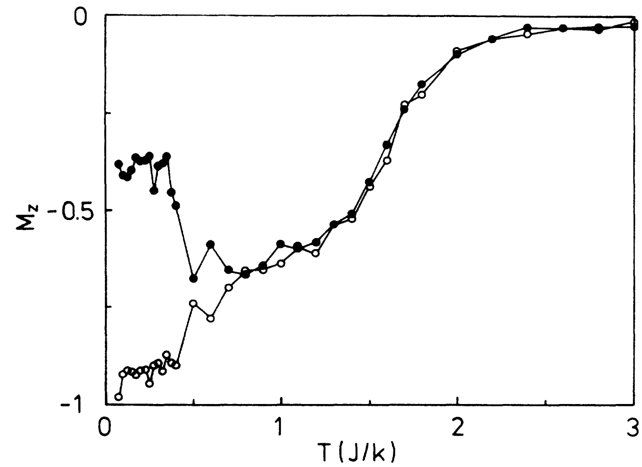

FIG. 3. Low-temperature magnetic moments computed for the sample with $N=1163$. The value of magnetic field chosen, $H=0.01\Phi_0/a_r^2$, approximately corresponds to its critical value for the sample of this size.

In summary, in our Monte Carlo simulations of the three-dimensional, strongly disordered system of Josephson junctions we see pronounced differences in the magnetic properties of the zero-field-cooled and field-cooled samples. These differences occur only for fields exceeding a certain critical value H_{c1}. Below this field we observe fully reversible behavior of the system discussed. Close to H_{c1}, at low temperatures many local magnetic moments change their polarities. For small samples this results in the paramagnetic overall behavior, and for the bigger one in the reduction of its diamagnetic response to the external field. This indicates that the role of this effect decreases when the system size is increased. Also, the value of H_{c1} decreases with the increasing size of our system.

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[1]R. A. Webb, F. Voss, G. Grinstein, and P. M. Horn, Phys. Rev. Lett. **51**, 690 (1983).

[2]M. Tinkham, D. W. Abraham, and C. J. Lobb, Phys. Rev. B **28**, 6578 (1983).

[3]R. K. Brown and J. C. Garland, Phys. Rev. B **33**, 7827 (1986).

[4]M. G. Forrester, Hu Jong Lee, M. Tinkham, and C. J. Lobb, Phys. Rev. B **37**, 5966 (1987).

[5]G. Deutscher, Y. Imry, and L. Gunther, Phys. Rev. B **10**, 4598 (1974).

[6]S. Teitel and C. Jayaprakash, Phys. Rev. B **37**, 598 (1982); Phys. Rev. Lett. **51**, 1999 (1983).

[7]C. J. Lobb, D. W. Abraham, and M. Tinkham, Phys. Rev. B **27**, 150 (1983).

[8]Sajeev John and T. C. Lubensky, Phys. Rev. Lett. **55**, 1014 (1985); Phys. Rev. B **34**, 4815 (1986).

[9]T. Halsey, Phys. Rev. Lett. **55**, 1018 (1985); Phys. Rev. B **31**, 5718 (1985); J. Phys. C **18**, 2437 (1985).

[10]J. G. Bednorz, M. Takashige, and K. A. Müller, Europhys. Lett. **3**, 379 (1987).

[11]Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).

[12]P. Chaudhuri, J. Mannhart, D. Dimos, C. C. Tsuei, J. Chi, M. Oprysko, and M. Schneuermann, Phys. Rev. Lett. **60**, 1653 (1988).

[13]R. Marcon, R. Fastampa, M. Giura, and C. Matacotta, Phys. Rev. B **39**, 2796 (1989).

[14]A. P. Malozemoff, L. Krusin-Elbaum, D. C. Cronmeyer, Y. Yeshurun, and F. Holtzberg, Phys. Rev. B **38**, 6490 (1988).

[15]J. Choi and J. José, Phys. Rev. Lett. **62**, 320 (1989).

[16]A. Majhofer, L. Mankiewicz, and J. Skalski, Phys. Rev. B **39**, 4332 (1989).

[17]We use a generalized version of the model considered by C. Ebner and D. Stroud, Phys. Rev. B **31**, 165 (1985).

[18]V. Ambegaokar and A. Baratoff, Phys. Rev. Lett. **10**, 486 (1963); **11**, 104 (1963).