Phenomenological Theory of the Paramagnetic Meissner Effect

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(Received 13 December 1993)

We study the paramagnetic Meissner effect in a network of Josephson junctions which include a random distribution of π junctions. We perform dynamical Langevin simulations for the field cooling (FC) and zero-field cooling (ZFC) susceptibilities. We show that while the ZFC susceptibility is diamagnetic and of the order of $-1/4\pi$, the FC susceptibility can be paramagnetic for a finite concentration of π junctions. The field and concentration dependence of the FC susceptibility is studied. The model presents some glassy properties and the results are in good qualitative agreement with recent experiments in ceramic Bi compounds.

PACS numbers: 74.25.Ha, 74.50.+r, 75.10.Nr

Since the discovery of the high-T_c superconductors, the ceramic structure of the first samples [1] renewed the interest in granular superconductivity, characterized by history-dependent magnetic properties [2–4]. Recently, Braunisch *et al.* [5] studied the magnetic response of ceramic samples of $Bi_2Sr_2CaCu_2O_8$ at very low fields. They found that, whereas the zero-field cooling (ZFC) experiment shows full flux expulsion, the field cooling (FC) susceptibility could be either much smaller than $-1/4\pi$ or even *paramagnetic.* In many samples the FC low-temperature susceptibility at very low fields (~ 0.1 Oe) is paramagnetic and large, typically 0.5 in units of $1/4\pi$. As the field increases the susceptibility decreases and becomes diamagnetic for fields larger than 0.5 Oe. Moreover, the temperature dependence of this so-called paramagnetic Meissner effect (PME) clearly indicates that this phenomenon is correlated with the occurrence of superconductivity. It has recently been proposed to call this phenomenon the Wohlleben effect [6].

A qualitative explanation for the PME based on the existence of spontaneous orbital currents was proposed in Ref. [5]. Kusmartsev [7] and Sigrist and Rice [6] independently studied the phenomenon assuming the existence of anomalous Josephson junctions between the grains. These anomalous junctions have a negative Josephson coupling, and they are called π junctions because the Cooper pairs acquire a phase π in the tunneling process. If in a loop of grains there is an odd number of π junctions, a spontaneous current could be generated giving rise to an orbital moment. A network with a concentration c of randomly distributed π junctions may behave as an orbital glass [7] and naturally account for the observed paramagnetic response. The microscopic origin of the π junctions is not clear. On one hand, Bulaevskii *et al.* [8] proposed some time ago that magnetic impurities in the junction can produce elastic tunneling associated with a spin flip process which induces an extra phase as Cooper pair tunnels. It has been argued, however, that in order to produce the observed effect, the impurity concentration must be rather large and interactions between impurities would suppress the effect. On the other hand, Sigrist and Rice interpreted the paramagnetic response of the Bi compounds as an indirect observation of d-wave superconductivity in this material. Experimental evidence of this has been reported recently by Wollman *et al.* [9] in dc-SQUID interferometry measurements.

Regardless of the origin of the π junctions, before accepting this description as the explanation of the PME, the complete thermodynamic behavior should be evaluated in a realistic model Josephson network in order to see if the temperature and field dependence of the FC and ZFC susceptibilities are consistent with the available experimental results.

Theoretically, granular superconductors have been modeled with networks where the nodes represent the grains and the links represent the coupling between grains through Josephson junctions [2–4]. In this work, we present the first simulation for the response of a network of Josephson junctions with a random distribution of π junctions. We show that this model, although rather simple, contains the essential ingredients for a quantitative description of the PME. We consider two dimensional (2D) and three dimensional (3D) networks. We include in our simulations the magnetic field induced by local screening current effects, which is crucial in order to obtain the PME.

The model studied in this paper is defined by the "coarse grained action"

$$\mathcal{F} = -\sum_{\mathbf{r},\hat{\mu}} J_{\mathbf{r}\hat{\mu}} \cos \Psi_{\hat{\mu}}(\mathbf{r}) + \frac{1}{2L} \sum_{\mathbf{R},\hat{\nu}} [\Phi_{\hat{\nu}}(\mathbf{R}) - \Phi_{\hat{\nu}}^{\text{ext}}]^2 . \quad (1)$$

The first term is the Josephson coupling energy, where $\Psi_{\hat{\mu}}(\mathbf{r}) = \theta(\mathbf{r}+\hat{\mu}) - \theta(\mathbf{r}) - \frac{2\pi}{\Phi_0}\int_{\mathbf{r}}^{\mathbf{r}+\hat{\mu}} \mathbf{A}\cdot d\mathbf{l}$ is the gauge invariant phase difference through the junction, with $\theta(\mathbf{r})$ the phase of the superconducting order parameter of the grain at $\mathbf{r}$, Φ_0 the flux quantum, and $\mathbf{A}$ the vector poten-

0031-9007/94/72(17)/2773(4)$06.00

tial. Here $(\mathbf{r}, \hat{\boldsymbol{\mu}})$ labels the junctions in a square (cubic) lattice which connect the grain lattice sites $\mathbf{r}$ and $\mathbf{r} + \hat{\boldsymbol{\mu}}$, with $\hat{\boldsymbol{\mu}}$ a unit vector along the x or y axis (or z axis). The Josephson coupling $J_{\mathbf{r}\hat{\mu}} = \pm \frac{\hbar I_c}{2e}$ is positive (negative) for 0 (π) junctions, with I_c the critical current of the junctions. In principle, each grain in the network has a given superconducting critical temperature T_{cg}, at which I_c vanishes. Thermal fluctuations of the superconducting phases $\theta(\mathbf{r})$ destroy the coherence between grains at a temperature T_c of the order of the Josephson energy $k_B T_c \sim \frac{\Phi_0 I_c}{2\pi}$. If the intergrain coupling is weak, i.e., I_c is small, T_c can be relatively small as compared with T_{cg}, and in the range $0 < T < T_c$ the temperature dependence of I_c can be neglected. For clarity, and in order to emphasize the effects of the phase fluctuations of the grains, we will work in this limit. The model has no topological disorder, and all the randomness is in the sign of $J_{\mathbf{r}\hat{\mu}}$.

The second term in Eq. (1) is the magnetic energy of the current induced magnetic fields. This term has been usually neglected in most of the simulations of Josephson networks [2,3], except in some recent 2D simulations [10]. The total magnetic flux through a given plaquette is given by $\Phi_{\hat{\nu}}(\mathbf{R}) = \Phi_{\hat{\nu}}^{\rm ext} + L I_{\hat{\nu}}(\mathbf{R})$, where $\Phi_{\hat{\nu}}^{\rm ext}$ is the flux per plaquette due to the external field, $I_{\hat{\nu}}(\mathbf{R})$ is the loop current, and L is the self-inductance of the loop [11]. Here $(\mathbf{R}, \hat{\boldsymbol{\nu}})$ labels the plaquette centered at the dual lattice site $\mathbf{R}$ and oriented along the direction $\hat{\boldsymbol{\nu}}$ (in 2D, $\hat{\boldsymbol{\nu}} = \hat{\mathbf{z}}$ only). The external magnetic field is along the z direction, $\Phi_{\hat{\nu}}^{\rm ext} = HS\hat{\mathbf{z}}$, with S the area of the plaquettes. The magnetic flux is related to $\Psi_{\hat{\mu}}(\mathbf{r})$ by $\frac{2\pi}{\Phi_0}\Phi_{\hat{\nu}}(\mathbf{R}) = -\Delta_{\hat{\mu}}^{\hat{\nu}} \wedge \Psi_{\hat{\mu}}(\mathbf{r})$, where the lattice curl operator $\Delta_{\hat{\mu}}^{\hat{\nu}}$ is, for example, $\Delta_{\hat{\mu}}^{\hat{z}} \wedge \Psi_{\hat{\mu}}(\mathbf{r}) = \Psi_{\hat{\mathbf{x}}}(\mathbf{r}) - \Psi_{\hat{\mathbf{x}}}(\mathbf{r}+\hat{\mathbf{y}}) + \Psi_{\hat{\mathbf{y}}}(\mathbf{r}+\hat{\mathbf{x}}) - \Psi_{\hat{\mathbf{y}}}(\mathbf{r})$. We consider thermal effects by studying the Langevin dynamical equations of motion for $\Psi_{\hat{\mu}}$,

$$\Gamma \frac{\partial \Psi_{\mu}(\mathbf{r}, t)}{\partial t} = -\frac{\delta \mathcal{F}}{\delta \Psi_{\mu}(\mathbf{r}, t)} + \eta_{\hat{\mu}}(\mathbf{r}, t) \; . \qquad (2)$$

The Langevin white noise $\eta_{\hat{\mu}}(\mathbf{r}, t)$ has correlations $\langle \eta_{\hat{\mu}}(\mathbf{r}, t)\, \eta_{\hat{\mu}'}(\mathbf{r}', t') \rangle = 2\Gamma k_B T \delta_{\mathbf{r},\mathbf{r}'} \delta_{\hat{\mu},\hat{\mu}'} \delta(t - t')$, where T is the temperature, k_B is Boltzmann's constant, and the dissipation parameter is $\Gamma = (\frac{\Phi_0}{2\pi})^2 \frac{1}{\mathcal{R}}$, with $\mathcal{R}$ the normal resistance of the links.

The system of differential equations given by Eq. (2) is integrated numerically with a second order Runge-Kutta algorithm suitable for stochastic systems [12]. Typical integration steps are $\Delta t = 0.02 - 0.1\tau_J$ ($\tau_J = \frac{\Phi_0}{2\pi \mathcal{R} I_c}$), and for each given temperature the integration is carried out for time intervals of $t = 3000\tau_J$, after an equilibration time of $750\tau_J$. To allow for the penetration of the magnetic field from the boundaries, we take free end boundary conditions.

We can simulate ZFC and FC experiments: at $T = 0$ the current distribution is calculated in the absence of an external field; this state is used as the initial condition to determine the stable state after turning on the external field. Then the temperature is increased by a small quantity δT always using the previous state as a seed to calculate the new current distribution, thus simulating a ZFC process. Once temperatures larger than T_c are reached, the temperature is slowly decreased with the field still on, simulating a FC process. At each temperature the magnetization $M(T, H, c)$ along the z direction is calculated as

$$m = \frac{4\pi M S}{\Phi_0} = \left\langle \frac{1}{N_z (N-1)^2} \sum_{\mathbf{R}} \frac{\Phi_{\hat{z}}(\mathbf{R}, t)}{\Phi_0} \right\rangle - f \; , \quad (3)$$

where $f = HS/\Phi_0$ is the normalized external field, and the network has $N \times N \times N_z$ grains ($N_z = 1$ in 2D). Here, $\langle \cdots \rangle$ is an average over time and over independent random configurations of π junctions. The normalized susceptibility is defined as $\tilde{\chi} = m/f = 4\pi\chi$, with $\chi = M/H$. At high temperatures the thermal noise produces large fluctuations in the physical magnitudes. The quality of the numerical results depends on the performed statistics which in large 3D networks may involve long CPU times, restricting us to take averages over a few disorder configurations.

We first study the effect of a single π junction at the center of a 2D square network of 0 junctions. The magnetic flux through each plaquette after a ZFC process at $T = 0$ is shown in Fig. 1(a). There is a current flowing through the π junction with the structure corresponding to a vortex and an antivortex pinned to this π defect. However, the magnetic flux in the two plaquettes next to the π junction is not one quantum flux but depends on the value of LI_c. This type of structure in the current distribution is characteristic of systems in which $\mathcal{L} = \frac{2\pi L I_c}{\Phi_0} > 1$; for small $\mathcal{L}$ the π junction induces no currents in its neighborhood. The whole π defect does not produce a net magnetization and the field expulsion is complete, as in a Meissner state. If we now increase the temperature above T_c, the magnetic field fully penetrates the sample. From this state, we decrease the temperature again to $T = 0$, as in a FC process. But now the field that has penetrated the sample polarizes the defect, giving rise to a new current distribution which resembles a vortex with the π junction in its core as shown in Fig. 1(b). In this state there is no current flowing through the π junction. As can be seen in the figure, in the FC state there are also other flux lines threading the sample, since even in a perfect network there is an intrinsic pinning [4] which prevents these other flux lines from flowing out of the sample. Therefore, after a FC process a π defect contributes with an additional positive magnetization. This same behavior of a single π junction occurs in 3D but with a nucleation of a vortex loop instead of a vortex-antivortex pair in the ZFC state.

The minimum magnetic field H^* necessary to polarize a π defect is related to the energy difference between the two current configurations induced by the defect. For the parameters corresponding to the figure, $H^* \approx 0.02\Phi_0/S$.

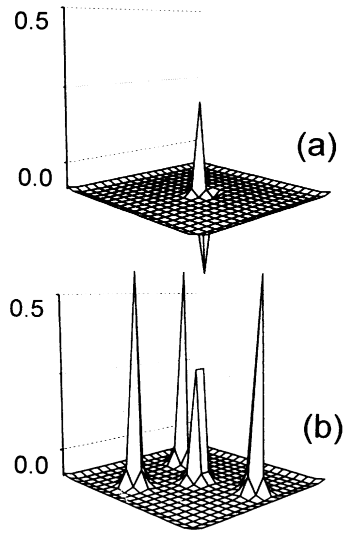

FIG. 1. Magnetic flux distribution $\Phi_{\hat{z}}(\mathbf{R})/\Phi_0$ of a two dimensional system of 20 × 20 sites with a π junction located at its center. Each point in the mesh represents the flux of a plaquette. The parameters are $\mathcal{L} = \frac{2\pi L I_c}{\Phi_0} = 8$ and the external field $H = 0.1\Phi_0/S$. The magnetization obtained in a ZFC process (a) and after a FC process (b) is shown.

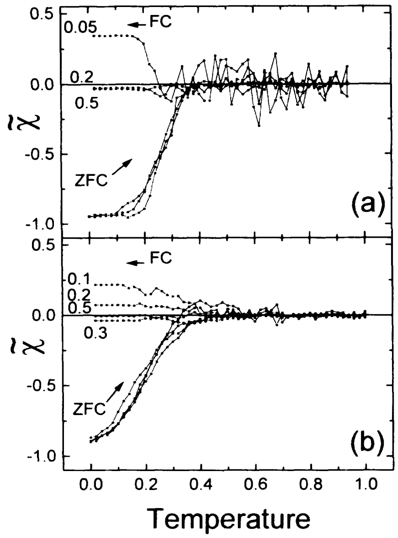

FIG. 2. Susceptibility $\tilde{\chi}$ as a function of temperature for lattices with $\mathcal{L} = 8$. (a) Two dimensional system of 16 × 16 sites with $c = 0.2$ and configurational average over three samples. (b) Three dimensional system of 8 × 8 × 8 sites with $c = 0.3$ and configurational average over four samples. The magnetic field in units of Φ_0/S is indicated. Temperature is in units of $\frac{\Phi_0 I_c}{2\pi k_B}$.

However, if in the same plaquette there are two parallel π junctions, the resulting defect has a similar structure with a threshold field H^* an order of magnitude smaller. A stair-shaped string of π junctions starting at the edge of the sample produces a single easily polarizable orbital moment at its end. In general, if there is a random distribution of π junctions there will be a distribution of fields H^*.

The ZFC and FC susceptibilities (χ_{ZFC} and χ_{FC}) can be calculated for an arbitrary concentration of π junctions. For the bipartite lattices we are studying, a system with a concentration c of randomly distributed π junctions can be mapped, through a gauge transformation, onto a system with a concentration $1 - c$. We then restrict to the case $c \leq 0.5$. Results for the susceptibility corresponding to a 2D network with $c = 0.2$ and a 3D network with $c = 0.3$, with high screening, $\mathcal{L} = 8$, and different values of the applied field, are shown in Figs. 2(a) and 2(b), respectively. In all cases the χ_{ZFC} at low temperatures is close to $-1/4\pi$. As the temperature increases it decreases and is zero for $T > T_c = \alpha\frac{\Phi_0 I_c}{2\pi k_B}$ where α is a numerical constant. From the figure we estimate $\alpha \sim 0.4$.

At low fields, the FC susceptibility increases with decreasing field and becomes positive, because of the contribution of all the polarized π junctions. This is the PME in the same way as it was seen in the experiments. In our simulations we found that for $\mathcal{L} < 1$, the PME is not clearly observed within the statistical error bars. This is also consistent with the experiments, where the PME is clearly seen only in samples with high critical currents (i.e., with high $\mathcal{L}$).

The experimental results show some peculiarities of the curves of magnetizations vs temperature close to T_c: the FC susceptibility starts being diamagnetic before becoming paramagnetic and, in some cases, a peak in the ZFC susceptibility as a function of T is also observed. We have reproduced these features for some parameter values although a detailed comparision with the experimental results is difficult due to the thermal noise in our simulations.

The FC susceptibility is positive for small fields and decreases as the field increases; it goes through a diamagnetic minimum and tends to zero at large fields as shown in Fig. 3(a). This same type of field dependence of χ_{FC} has been observed experimentally. The FC susceptibility depends also on the concentration of π junctions in the sample. In Fig. 3(b) we show this dependence for a 3D lattice at a fixed external field. The concentration dependence of the FC susceptibility can be estimated by counting the number of plaquettes having an odd number of π junctions, and thus giving a paramagnetic contribution to χ_{FC}. In the figure we show a fitting of the numerical data calculated in this way, the only free parameter being the effective susceptibility of each one of these plaquettes.

Even when $\chi_{\text{FC}} > 0$, the differential susceptibility is always diamagnetic. If we are in the FC curve with $\chi_{\text{FC}} > 0$ at a given temperature and slightly increase (decrease) the external field, the magnetization linearly decreases (increases).

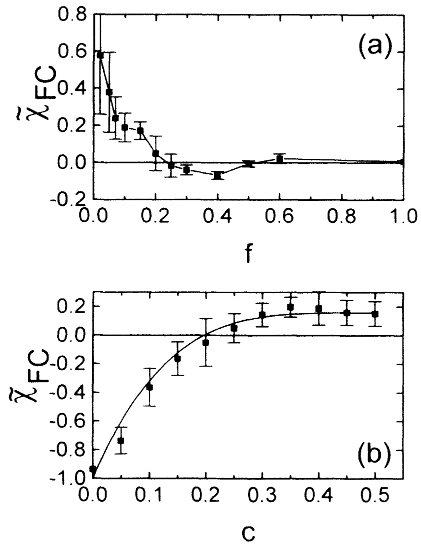

FIG. 3. FC susceptibility $\tilde{\chi}_{FC}$ for $8 \times 8 \times 8$ samples with $\mathcal{L} = 8$, obtained with a cooling rate of $\delta T = -0.02\frac{hI_c}{2ek_B}$ and averaged over five samples. (a) As a function of field $f = HS/\Phi_0$ for concentration $c = 0.3$. (b) As a function of concentration for $f = 0.1$. In (b) the solid line shows the fitting described in the text.

Although the general behavior of these systems is independent of the cooling rate δT, the low-temperature value of χ_{FC} depends on it. In general we obtain that a smaller δT gives rise to a larger χ_{FC}. This dependence on the cooling rate is characteristic of spin glasses [13]. In fact, the model of Eq. (1) is an XY spin glass [13] but with a particular quadratic coupling with the magnetic field. Thus, the picture of an orbital glass seems to be more appropriate than the simple model of independent loops analyzed in Ref. [6]. A complete study of the glassy properties of the model would require the calculation of the relaxation rates at a fixed temperature and field. This problem, which presents technical difficulties since it involves long simulation times, is presently under study.

In summary, we have calculated the field and temperature dependence of the magnetization for a network of Josephson junctions with a random distribution of anomalous π junctions. The results clearly show that while the low-temperature ZFC susceptibility at small or moderate values of the external fields is of the order of $-1/4\pi$, the FC susceptibility is paramagnetic for certain parameter values. The paramagnetic response after a FC process is obtained for a finite concentration of π junctions, large self-inductances or critical currents, ($\frac{2\pi LI_c}{\Phi_0} > 1$) and a small external field ($H \ll \Phi_0/S$). The field dependence of χ_{FC} at low temperatures is in agreement with the experimental results. The model presents some characteristics of a spin glass system, in particular the dependence of the low-temperature value of χ_{FC} with the cooling rate. Our results are in general in good agreement with the experimental data and support the idea of an orbital glass description of the Bi-2:2:1:2 compounds.

We would like to acknowledge helpful discussions with D. Khomskii and F. de la Cruz. E. A. J. is supported by CONICET, and C. A. B. is partially supported by CONICET.

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[1] K. A. Muller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. **58**, 1143 (1987).
[2] W. Y. Shih, C. Ebner, and D. Stroud, Phys. Rev. B **30**, 134 (1984); C. Ebner and D. Stroud, Phys. Rev. B **31**, 165 (1985); S. John and T. C. Lubensky, Phys. Rev. B **34**, 4815 (1986).
[3] I. Morgenstern, K. A. Muller, and J. G. Bednorz, Z. Phys. B **69**, 33 (1987); J. Choi and J. V. José, Phys. Rev. Lett. **62**, 320 (1989); G. Ramírez-Santiago and J. V. José, J. Phys. A **26**, L535 (1993); Y. H. Li and S. Teitel, Phys. Rev. Lett. **66**, 3301 (1991).
[4] M. Tinkham and C. J. Lobb, in *Solid State Physics*, edited by H. Ehrenreich and D. Turnbull (Academic, San Diego, 1989), Vol. 42, p. 49.
[5] W. Braunisch *et al.*, Phys. Rev. Lett. **68**, 1908 (1992); Phys. Rev. B **48**, 4030 (1993).
[6] M. Sigrist and T. M. Rice, J. Phys. Soc. Jpn. **61**, 4283 (1992).
[7] F. V. Kusmartsev, Phys. Rev. Lett. **69**, 2268 (1992).
[8] L. N. Bulaevskii, V. V. Kuzii, and A. A. Sobyanin, Pis'ma Zh. Eksp. Teor. Fiz. **25**, 314 (1977) [JETP Lett. **25**, 290 (1977)].
[9] D. A. Wollman *et al.*, Phys. Rev. Lett. **71**, 2134 (1993).
[10] A. Majhofer, T. Wolf, and W. Dieterich, Phys. Rev. B **44**, 9634 (1991); D. Domínguez and J. V. José, Phys. Rev. Lett. **69**, 514 (1992); J. R. Phillips *et al.*, Phys. Rev. B **47**, 5219 (1993).
[11] We do not consider mutual inductances between loops, which only renormalize the effective penetration depth of the 3D network, without affecting the long-range interaction between vortices (however, the same is not true for 2D; see Ref. [10]).
[12] H. S. Greenside and E. Helfand, Bell. Syst. Tech. J. **60**, 1927 (1981).
[13] K. Binder and A. P. Young, Rev. Mod. Phys. **58**, 801 (1986); S. Jain and A. P. Young, J. Phys. C **19**, 3913 (1986).