Reentrant ac Magnetic Susceptibility in Josephson-Junction Arrays

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(Received 19 February 1997)

We have measured the complex ac magnetic susceptibility of unshunted Josephson-junction arrays as a function of temperature T, amplitude of the excitation field h_{ac}, and external magnetic field H_{dc}. For small h_{ac} Meissner screening occurs. For larger h_{ac}, however, the screening is reentrant in T. This reentrance is not thermodynamic but dynamic and arises from the paramagnetic contribution of multijunction loops. This result gives an alternative explanation of the paramagnetic Meissner effect observed in granular superconductors. Experimental results are in agreement with a simplified model based on a single loop containing four junctions. [S0031-9007(97)03358-9]

PACS numbers: 74.50.+r, 74.25.Ha, 74.80.Bj

A paramagnetic Meissner effect (PME) has been measured in Bi-based high-T_C granular superconductors [1,2]. This effect has been attributed to the presence of π junctions between the grains. In these junctions, the Cooper pair acquires a phase shift π across the junction, giving rise to Josephson currents which are *negative* relative to conventional junctions [3]. Such a phase shift might result from magnetic impurities between the grains or non-s-wave pairing symmetry [4,5].

This Letter reports the appearance of a strong paramagnetic contribution to the complex ac magnetic susceptibility of niobium Josephson junction arrays. Since our arrays are made of conventional junctions, our result shows that PME can occur without the presence of π junctions. In our experiment, the paramagnetic contribution appears as a reentrant behavior of the ac suceptibility, χ_{ac}, at low temperature. The in-phase component of the first harmonic, χ_1', which is a measure of the screening current, first increases in modulus as the temperature is lowered from the critical temperature T_C, then decreases at a lower temperature. The out-of-phase component, χ_1'', is correlated with χ_1', showing increasing losses as the screening decreases at low temperature (see Fig. 1) [6]. Moreover, we find that numerical simulations of the simplified case of a four junction loop exhibit paramagnetic susceptibility in some ranges of excitation field and temperature, accounting very satisfactorily for our experimental results.

Our arrays have square geometry and consist of unshunted Nb-AlO$_X$-Nb tunnel junctions (see Fig. 2). The lattice spacing is $a = 46\ \mu$m; from the dimensions of the films, we estimate that the inductance of each loop is about $L = 64\ p$H. The critical current density of the junctions is about 600 A/cm^2 at 4.2 K, and the junction area is $5 \times 5\ \mu\text{m}^2$. We performed measurements of χ_{ac} as a function of the temperature T ($1.5 < T < 15$ K), the amplitude of the excitation field h_{ac} (0.5 mOe $< h_{ac} <$ 10 Oe), and the external dc field H_{dc} ($0 < H_{dc} <$ 700 Oe) parallel to the plane of the sample. We use a homemade susceptometer in a screening configuration [13]. The susceptometer is positioned inside a double wall μ-metal shield, screening the sample from the Earth's magnetic field.

Data for χ_1' and χ_1'' as a function of temperature are shown in Fig. 1. For small values of h_{ac}, the behavior of both components of χ_{ac} is quite similar to that found in superconducting samples [14], i.e., χ_1' becomes more negative at lower temperatures, indicating stronger superconductivity through the Meissner effect, and χ_1'' peaks, indicating a maximum in the losses, around the critical temperature T_C. Remarkably, however, for values of h_{ac} larger than about 50 mOe, the in-phase component, χ_1', is reentrant. χ_1'' is correlated with the reentrance observed in χ_1', showing increasing losses as the screening decreases, indicating an apparent weakening of superconductivity at low temperatures. The minimum in χ_1' appears at $T \simeq 7.0$ K.

The ac response of two-dimensional arrays has been a powerful tool in studying phase transitions [15]. It is tempting to identify the reentrance with a phase transition. For example, the array might become ordered at $T \simeq 9$ K [16], and then become disordered again at $T \simeq 7.0$ K. Such reentrance was predicted to occur in disordered arrays

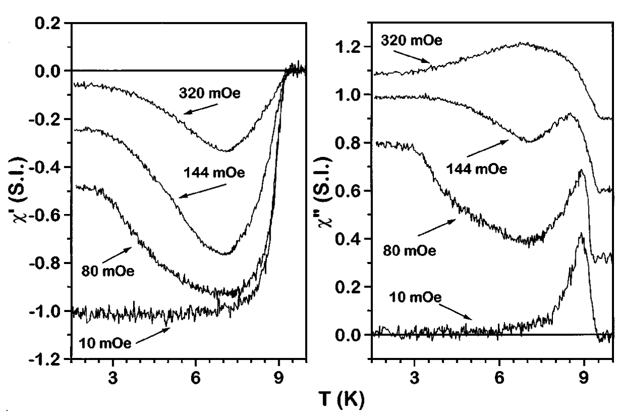

FIG. 1. χ_1' and χ_1'' as a function of T for different values of h_{ac} with $H_{dc} = 0$. The curves for χ_1'' have been vertically offset for clarity.

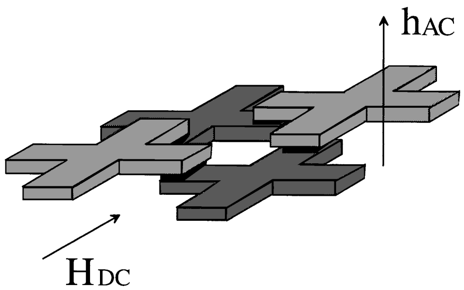

FIG. 2. Sketch of a small section of a sample. The crosses are the niobium islands. The junctions are in the overlap region between these islands.

in a perpendicular magnetic field, but experiments and more recent theory question this prediction [17]. The array studied here is, however, very strongly coupled, with a ratio of the coupling energy to the temperature $\Phi_0 I_C/2\pi kT \simeq 260$ at $T = 7.0$ K, where Φ_0 is the magnetic flux quantum. A reentrant phase transition therefore seems unlikely. As an alternative, we examine the dynamics of the screening currents.

In order to investigate the origin of the reentrance, we exploited a feature of our array design. Figure 2 shows that a magnetic field applied parallel to the plane of the substrate will suppress the critical current of each junction by causing flux to penetrate each junction without introducing any flux into the unit cells (or plaquettes) of the array. (A perpendicular field introduces flux in the "holes" in each unit cell.) Thus, a parallel magnetic field allows us to vary I_C independently from temperature or the applied perpendicular field.

Figure 3 shows the results of measurements of $\chi_{\rm ac}$ as the parallel field (and thus the critical current) is varied. The minimum in χ_1' is shifted to lower temperatures by this, consistent with the weakening of the critical current and thus of the maximum screening current. Furthermore, we find that χ_1' (4.2 K) shows the same Fraunhofer dependence on $H_{\rm dc}$ as the critical current I_C of the junctions forming the array, giving further proof that only the critical current is varied in this experiment.

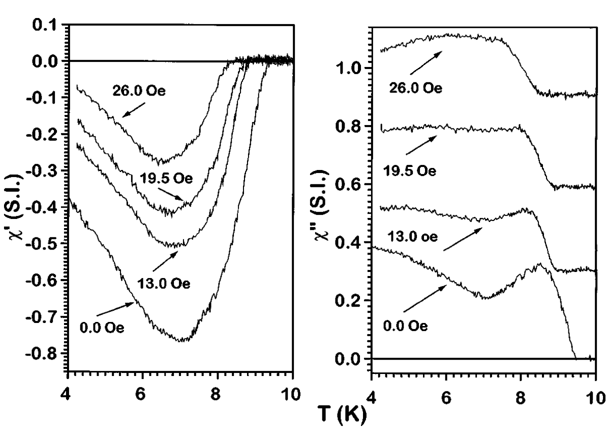

FIG. 3. χ_1' and χ_1'' vs T for $h_{\rm ac} = 144$ mOe and different values of $H_{\rm dc}$. The curves for χ_1'' have been vertically offset for clarity.

Although a complete explanation for the reentrance undoubtedly requires that the full array be considered, we have found that a simple model based on one unit cell of the array—a loop including four junctions—gives a good semiquantitative description of the reentrance. A similar model (consisting of a single-junction loop) has been previously used by Auletta *et al.* [18] to suggest a possible explanation of the PME in high T_C granular superconductors [1,2], by simulating the field-cooled dc magnetic susceptibility.

In a single loop,

$$\Phi_{\rm TOT} = \Phi_{\rm EXT} + LI, \tag{1}$$

where I is the circulating current in the loop and $\Phi_{\rm TOT}$ and $\Phi_{\rm EXT}$ are the total flux and the flux related to the applied magnetic field, respectively. The junctions are modeled by taking into account their capacitance C_J and their quasiparticle resistance R_J. Therefore, the current I is given by

$$I = I_C \sin\gamma_i + \frac{\Phi_0}{2\pi R_J}\frac{d\gamma_i}{dt} + \frac{C_J\Phi_0}{2\pi}\frac{d^2\gamma_i}{dt^2}, \tag{2}$$

where γ_i is the superconducting phase difference across the ith junction and I_C is the critical current of each junction. In the case of four junctions, the fluxoid quantization condition, which relates each γ_i to the external flux, is

$$\gamma_i = \frac{\pi}{2}n - \frac{\pi}{2}\frac{\Phi_{\rm TOT}}{\Phi_0}, \tag{3}$$

where n is an integer and, by symmetry, we assume $\gamma_1 = \gamma_2 = \gamma_3 = \gamma_4 = \gamma_i$. In the case of an oscillating external magnetic field, $H_{\rm EXT} = h_{\rm ac}\cos(\omega t)$, the magnetization $M = LI/\mu_0 a^2$, where μ_0 is the vacuum permeability, may be expanded as a Fourier series,

$$M(t) = h_{\rm ac}\sum_{n=0}^{\infty}[\chi_n'\cos(n\omega t) + \chi_n''\sin(n\omega t)]. \tag{4}$$

In the model, the temperature dependent parameter is the critical current of the junctions. We used the approximation [19]

$$I_C(T) = I_C(0)\sqrt{1 - T/T_C}\,\tanh[1.54T_C\sqrt{(1 - T/T_C)}/T]. \tag{5}$$

We have performed numerical simulations based on Eqs. (1)–(5) and calculated the zero field-cooled χ_1 as a function of T for different amplitudes of $h_{\rm ac}$. Figure 4 shows a plot of these results.

The simulated χ_1' is qualitatively very similar to the experimental data, showing reentrance at low temperature. The shape of χ_1' depends on the parameter $\beta_L(T) = 2\pi I_C(T)L/\Phi_0$, which is proportional to the number of

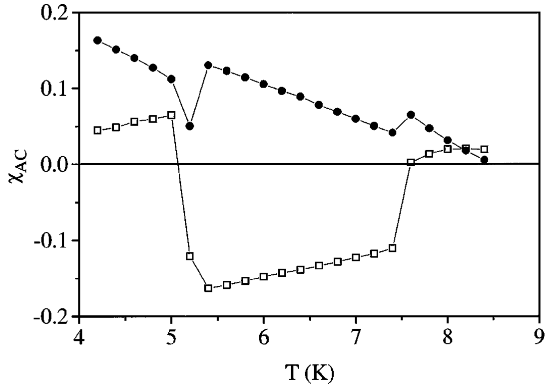

FIG. 4. Simulation of χ_1 vs T for $\Phi_{\mathrm{EXT}}/\Phi_0 = 7$ and $\beta_L(4.2\ \mathrm{K}) = 30$. χ_1' is indicated by open squares and χ_1'' by solid circles.

flux quanta that can be screened by the critical current in the junctions. For our array, $\beta_L(4.2\ \mathrm{K}) = 30$. The curve Φ_{TOT} vs Φ_{EXT} is very hysteretic, showing multiple branches (see Fig. 5). At each temperature, the branch that intersects the line $\Phi_{\mathrm{TOT}} = 0$ corresponds to diamagnetic states. For all the other branches, their intersection with the line $\Phi_{\mathrm{TOT}} = \Phi_{\mathrm{EXT}}$ corresponds to the boundary

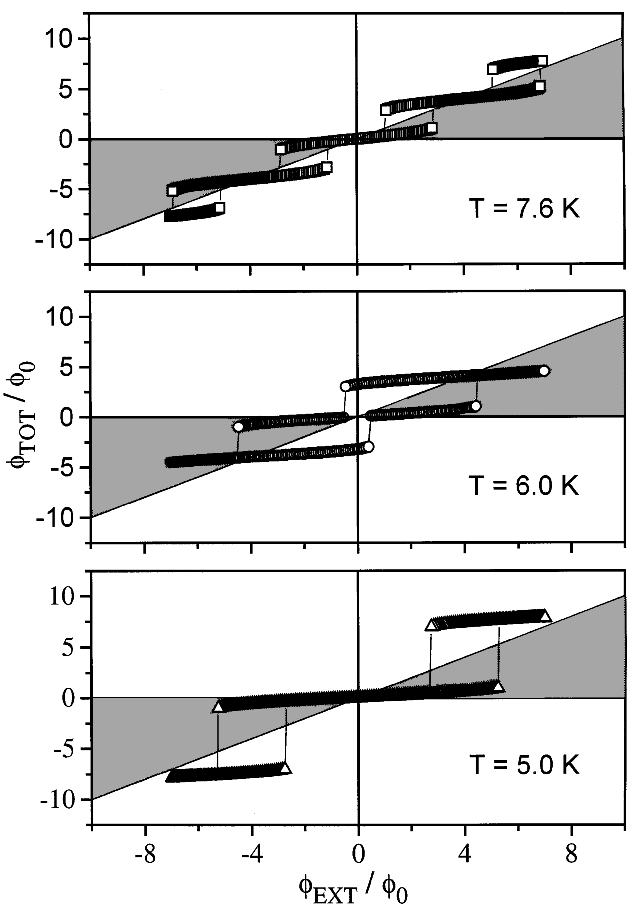

FIG. 5. Simulated total flux versus external flux at different temperatures.

between paramagnetic states (positive χ_1') and diamagnetic states (negative χ_1'). In Fig. 5 the shaded areas correspond to diamagnetic states ($\Phi_{\mathrm{TOT}} < \Phi_{\mathrm{EXT}}$) and the clear areas to paramagnetic states ($\Phi_{\mathrm{TOT}} > \Phi_{\mathrm{EXT}}$). The value of χ_{ac} is given by an average among all the states that the system traverses in one cycle of the ac drive. Therefore, χ_{ac} will be paramagnetic or diamagnetic, depending on the segments of the hysteretic curve which are spanned during the cycle.

The shape of the curve changes with temperature, as is shown in Fig. 5. The crossover from paramagnetic to diamagnetic behavior can be understood by looking at simulations showing Φ_{TOT} vs Φ_{EXT} at different temperatures. At the temperature values $T = 5$ K and $T = 7.6$ K, the appearance of the second and third branches, respectively, add a paramagnetic contribution to the average value of χ_1', thus explaining the sharp changes of χ_1' vs T shown in Fig. 4.

The response of the array presents no sharp transitions because it is the result of an average response from all the loops, and also depends on the profile of the field penetration in the whole array [20–24]. Moreover, in order to completely account for the measured χ_{ac} we should also consider the additive diamagnetic response of the Nb islands.

Our simplified description of the system qualitatively accounts for all the experimental observations. The reentrance in χ_1' is due to the paramagnetic contribution of the multijunction loops. When the dimensionless ratio $\mu_0 h_{\mathrm{ac}} a^2/LI_C$ is large enough, the system switches to the next branch of Φ_{EXT} vs Φ_{TOT} curve (see Fig. 5), leading to a paramagnetic component of the response, which reduces the overall diamagnetic response. Similarly, each time the system switches from one branch to the other, an energy of the order Φ_0^2/L is dissipated, so that extra dissipation results when the system switches from a single-branch to a multibranch solution, i.e., when the solution becomes hysteretic. This argument explains the increased dissipation, proportional to χ'', measured at low temperature. Moreover, the values of χ_1' and χ_1'' for a fixed h_{ac} are approximately constant below $T \simeq 3$ K (see Fig. 1), in agreement with the fact that I_C is approximately constant (saturating to its maximum value) in that range of temperature.

In our experiment, this reentrance appears for values of h_{ac} higher than about 50 mOe, in good agreement with our estimated value of $LI_C/\mu_0 a^2 = 3.7$ A/m $= 47$ mOe. We conclude that the reentrance is associated with a dynamic mechanism and not a thermodynamic one.

Reentrant susceptibility has not been observed in granular superconductors, which are very similar to Josephson junction arrays [19,25–28]. However, granular superconductors have a distribution of critical currents and loop sizes, and thus a distribution of β_Ls. Therefore, for some fixed value of h_{ac}, some loops will remain diamagnetic while others will become paramagnetic at different temperatures. This is a possible explanation for the well-known intergrain plateau, a region of approximately constant value of χ_1' often measured in granular superconductors [14]. In fact, typical values of β_L in granular high T_C superconductors are in the range 5–200 [29]. Therefore, the phenomena related to the reentrance we have observed in Josephson junction arrays, where $\beta_L = 30$, should exist in granular systems.

Our experimental result shows that the multijunction loop model can also explain the PME effect observed in some field-cooled high T_C superconductors, in agreement with the simulations by Auletta *et al.* [18]. Experiments show that PME occurs when some weak links have a sufficiently large critical current, therefore the effect occurs above a corresponding minimum value of β_L. This may indicate that when PME occurs, the curve Φ_{EXT} vs Φ_{TOT} corresponding to some of the weak link loops is very hysteretic. When the samples are zero field cooled and then measured at small fields, most of the loops will be in states corresponding to the diamagnetic branch crossing $\Phi_{\text{TOT}} = 0$ and no paramagnetic response can be measured. When the samples are field cooled in small fields, flux quanta get trapped in the loops, corresponding to states in upper overlapping branches. These branches are paramagnetic at small values of field, and become diamagnetic at higher field. This explains the measured crossover from paramagnetic to diamagnetic response by increasing the field. Further increase of the dc field will eventually substantially reduce the critical current of the weak links, then only the diamagnetic response of the grains will contribute to the measured value of susceptibility. Further investigations of this possible explanation for the observed PME in high T_C superconductors requires a characterization of the samples and an estimation of the average β_L. We also note that PME has been recently measured in niobium disks [30]. Analogously to our result, this experiment confirms that PME also arises from trapped flux and that it is not necessarily related to d-wave symmetry.

We thank M. G. Forrester, A. W. Smith, and C. B. Whan for their technical help in the experiments. We also thank P. Fournier, R. L. Greene, R. Newrock, W. A. Ortiz, and F. Wellstood for useful discussions. We gratefully acknowledge financial support from U.S. Air Force Office of Scientific Research, through Grant No. F49620-92-J-0041, and from NSF through Grant No. 9510464. Also, F. M. A. M. thanks Brazilian Agencies FAPESP (Grant No. 96/7704-6) and CNPq (Grant No. 201328/91-7) for financial support.

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