Observation of Spontaneous Flux Generation in a Multi-Josephson-Junction Loop

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(Received 19 August 1999)

We describe observations of spontaneous flux generation inside a $YBa_2Cu_3O_{7-\delta}$ loop made of 214 Josephson junctions in series. The flux is generated spontaneously during cooldown into the superconducting state. The experiment is motivated by the Kibble-Zurek scenario of formation of topological defects in condensed matter systems. The transition from decoupled superconducting segments into a coherent loop is determined by the strength of thermal fluctuations in the junctions. Values of the flux measured at the end of each cooldown follow a normal distribution, and are consistent with the instantaneous phase differences across the junctions adding up as the loop becomes coherent.

PACS numbers: 74.50.+r, 03.75.Fi, 11.15.Ex, 74.40.+k

Some years ago, Zurek [1] proposed that condensed matter systems having a complex order parameter can be used to test a certain class of early universe theories discussed by Kibble and co-workers [2,3]. It was suggested that during the transition to an ordered state, while the system is out of equilibrium, separate regions are formed having random values of the phase of the order parameter. If such regions become connected in the form of a loop, and the sum of the phase differences exceeds 2π, a topological defect is created [1,2,4]. For example, in a superconductor this implies that there will be a spontaneous supercurrent around the loop, and trapped flux. The discussion as to whether this scenario is viable was linked to the basic question of the meaning of the phase of the order parameter [4–6], especially in the case where several unconnected regions exist. Obviously, the confirmation of this scenario would be of great importance. Among the candidates predicted to show this behavior are superconductors, superfluids, or Bose-Einstein condensates. However, experimental tests of this hypothesis on bulk systems gave conflicting results. Spontaneous nucleation of topological defects following a quench was observed in liquid crystals (disclinations) [7] and in superfluid ^{3}He (vortices) [8,9], but not in ^{4}He [10]. Similarly, no spontaneous flux was detected in a bulk high temperature superconductor [11]. In order to try to resolve this situation, it is important to test the very basis of this hypothesis, at the level of what happens in a single loop. To that end, we designed such an experiment to search for spontaneous flux generation in a single loop composed of many superconducting segments having random phase differences, which then become connected.

In our experiment we use a $YBa_2Cu_3O_{7-\delta}$ (YBCO) loop, interrupted by 214 grain-boundary Josephson junctions in series. The loop is patterned from a 700 Å epitaxial c-axis-oriented YBCO film, grown on 24° $SrTiO_3$ bicrystal substrate, 1×1 cm^2 size (for properties of grain boundaries in YBCO, see Refs. [12,13]). The pattern, shown in Fig. 1, is a 20 μm wide meander line. Each time this line crosses the grain boundary, another Josephson junction is added [14]. The area of the loop is 0.3 cm^2. The number of junctions is limited by the maximal number of 20 μm-wide segments we can insert along the grain boundary line.

The experimental setup and the measuring method are basically the same as described previously [11]. Briefly, in order to measure the flux threading the loop we use a high temperature SQUID magnetometer placed close to the sample. The sample is heated above T_c (= 90 K), using a light beam, and is cooled via a thermal link to liquid nitrogen through helium exchange gas. In this way we avoid any stray magnetic fields due to electrical currents driving a conventional heater. The SQUID is maintained at a constant temperature of 77 K, independent of the sample. The noise level of the SQUID corresponds to an uncertainty of $\pm 1.5\phi_0$ inside the loop. The external magnetic field at the sample was shielded to better than 10^{-4} G. This residual field could be varied using a small *in situ* coil.

By using the SQUID, a continuous measurement of the flux through the loop is performed while cooling the sample from $\simeq$100 to 77 K. At T_c(= 90 K), segments of

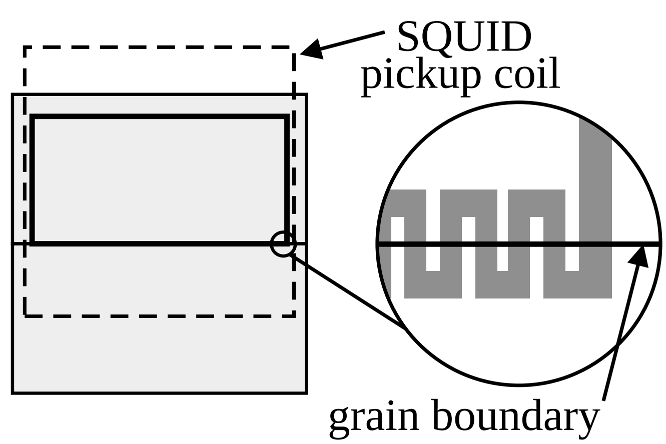

FIG. 1. A schematic representation of the loop pattern on the bicrystal substrate (solid line). The dashed line outlines the input coil of the SQUID. The enlarged area shows the superconducting meander line crossing the grain boundary, forming Josephson junctions in the process.

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the film separating the junctions become superconducting. However, the junctions are still normal and thus the superconducting segments are effectively separate. As the temperature decreases, the critical currents of the junctions increase, and eventually the loop becomes coherent. Indeed, the measurements show that spontaneous flux is trapped in the loop. A series of such measurements is shown in the top part of Fig. 2. For clarity, we display the final part of each measurement, where the temperature is already stable and flux inside the loop is constant (we actually continue the measurement for a much longer time to ascertain that). The sign and magnitude of the flux appear random from one cooldown to the next. The bottom part of Fig. 2 shows control experiments, done in exactly the same way, except that a blank substrate was used instead of the superconducting loop. The control experiment clearly shows that the trapped flux has nothing to do with residual field noise. We repeated the measurements under different residual fields (10^{-2} to 10^{-4} G) and at different cooling rates (20 to 0.3 K/sec), and found that the data were independent of both these factors over 2 orders of magnitude.

The distance between the loop and the SQUID is 1 mm. The measured coupling ratio of flux inside the loop to the SQUID is 0.37. If there are some vortices pinned inside the film itself or in the junctions, their coupling to the SQUID will be much smaller because the field lines will close around the superconducting strip, at a typical distance comparable to the width (20 μm), i.e., much less then the distance to the SQUID. Thus, we are only sensitive to the flux which is inside the loop.

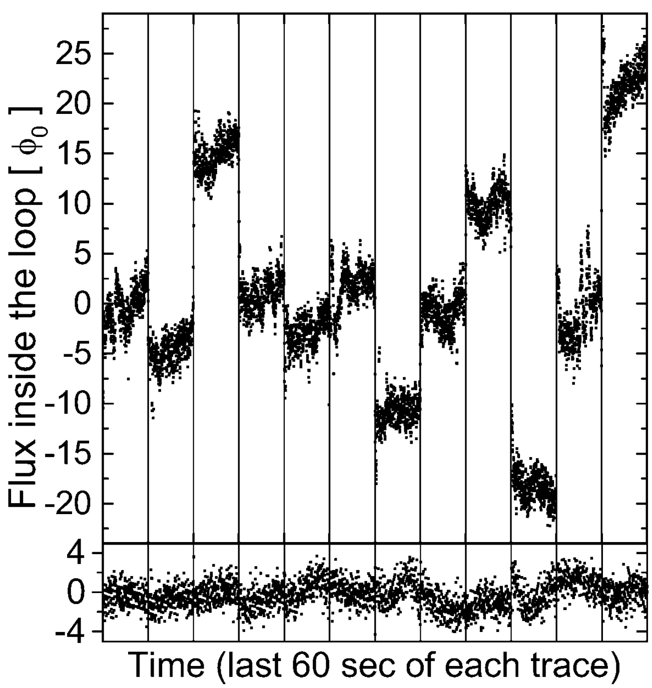

FIG. 2. The top part shows a typical sequence of stable values of the spontaneous flux in the loop measured by the SQUID during several consecutive cooling cycles. The bottom part shows identical reference measurements with a blank substrate.

In Fig. 3 we show the distribution of the stable flux levels inside the loop. The histogram contains the data obtained during 166 cooldowns. The standard deviation of the distribution is $7.4 \pm 0.7\phi_0$. The error bars here refer to the uncertainty in the absolute calibration of the flux sensitivity of our system. The solid line is a fit of the data to a normal distribution.

The specific properties of our system differ in several respects from that of the generic Kibble-Zurek scenario. However, the basic concept of linking segments with random phases is tested in our experiment. The Kibble-Zurek mechanism requires the freezing of the value of the cumulative phase difference around the loop by means of a thermal quench through the critical region. In our case, we propose that the total phase difference around the loop reflects the thermal fluctuations of the phase differences across the junctions at the time the loop becomes coherent. In the original Kibble-Zurek mechanism the cooling rate of the sample is crucial since it affects the value of the freeze-out coherence length (the size of the segments). In our experiment the physical size of the segments is constant ($\simeq$60 μm). Each segment reaches internal equilibrium long before the junctions. Thus, the cooling rate should not matter in our setup, as indeed observed. Thermal fluctuations in grain boundary junctions were discussed in several publications [12,15]. During cooldown, the segments between adjacent junctions become superconducting at T_c while the maximum Josephson current I_c, is still small. At this stage, the coupling energy of the junction, $E_J \equiv I_c\phi_0/\pi$ is much smaller than k_BT. The various segments are effectively uncoupled and the loop is incoherent. In the original scenario, this situation is equivalent to a loop made

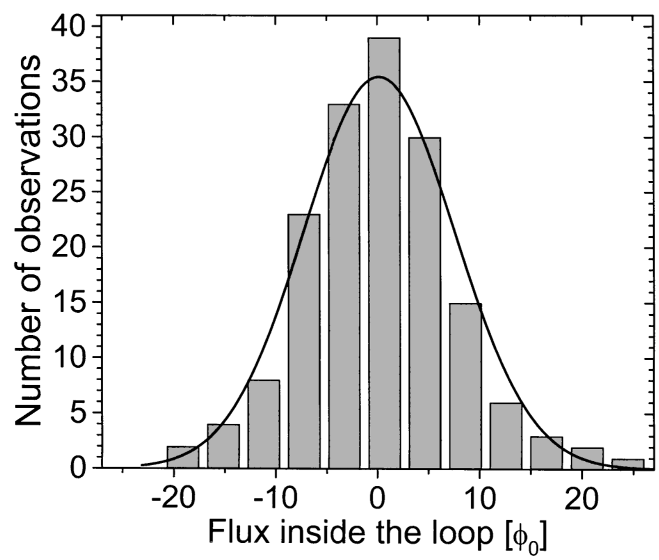

FIG. 3. The distribution of the values of spontaneous flux inside the loop, taken from 166 cycles. Values falling within $\pm 1.5\phi_0$, i.e., within the noise level of the SQUID, are binned together. The solid line is a fit of the data to normal distributions with standard deviation of $7.4\phi_0$.

up of bubbles which are yet unconnected. Thermal fluctuations of individual junctions are described by a probability to have a random phase difference δ_i given by $P(\delta_i) \sim \exp[-I_c\phi_0 \times (1 - \cos\delta_i)/(2\pi k_B T)]$, where i refers to the ith junction [12,16] (here δ_i is the gauge invariant phase difference). Any time a fluctuation occurs changing δ_i by $\geq 2\pi$, the whole loop becomes incoherent, meaning that the topological charge has changed (the maximal energy barrier corresponds to a change by π, and a change larger than 2π "opens" the loop). At this stage the fluctuating supercurrent through each of the junctions will be balanced locally by a reverse normal current, and there is no supercurrent flowing around the loop. As the temperature decreases, I_c increases and with it the coupling energy. Individual fluctuations still exist, but δ_i is less likely to jump by 2π . At this stage, the loop becomes coherent and a supercurrent can flow around the loop. While the loop is closed, small thermal fluctuations do not change the topological charge of the loop. As the loop cools, it closes and opens many times. Experimentally, this manifests itself as flux noise.

The loop can be considered as finally closed if the probability for δ_i to jump by 2π during the time interval of the measurement (typically, 1 min) is small. A lower limit on the time interval between such events is $\hbar/k_B T \times \exp[I_c\phi_0/\pi k_B T]$. This time interval reaches 1 min once $I_c \simeq 60\ \mu$A ($E_J \simeq 30 k_B T$). Typically, I_c of junctions such as used here reaches 60 μA, 5–7 K below T_c (I_c cannot be measured directly in our setup, but we do see a significant reduction of the measured flux noise during cooldown in this range of temperatures, which indicates that the loop indeed is closed). In the closed state, the total flux $\Phi = LI$, where L is the inductance of the loop and I is the supercurrent. The flux Φ is given by the sum of all of the phase differences around the loop $\Phi \simeq \phi_0 \sum_i \delta_i/2\pi$. It is important to emphasize that if the loop becomes coherent via sequential locking of more and more junctions, in the end it will reach the true ground state of the system with zero flux. Similarly, a mechanism of thermally activated trapping of flux inside the loop via single flux quanta passing through individual junctions (during a possible equilibrium cooling process) should leave no flux at the end of the cooldown since the loop will decay into the ground state. Only the presence of thermal fluctuations in *all* of the junctions enables the existence of random phase differences at the moment the loop becomes closed, and, hence, a finite flux.

The time scale for a flux quantum to pass through a junction is $\tau_J \approx 0.3\phi_0/R_J I_c$ ($R_J \approx 1\ \Omega$ is the normal resistance of a junction) [16]. In our case $\tau_J \approx 10^{-11}$ sec, at $I_c \simeq 60\ \mu$A. The time it takes the order parameter to adjust across the whole loop in this range of temperature is of the order $\tau_{\rm op} = (1\ {\rm cm}) \times \tau/\xi$ (the speed of the order parameter is ξ/τ, with the coherence length $\xi = 16$ Å and the relaxation time of the order parameter $\tau \simeq 10^{-12}$ sec). We find $\tau_{\rm op} \approx 10^{-5}$ sec. Finally, there is the electric time constant for closing the loop, L/R_T, where R_T is the series resistance of the junctions which are still normal at that instant (we estimate $R_T \approx 10\ \Omega$). For our system, $L \simeq 4\pi \times 10^{-9} r \ln(r/a)$ H, the radius $r \simeq 0.3$ cm and the width $a \simeq 2 \times 10^{-3}$ cm, yielding $L \simeq 20$ nH. The value of L/R_T comes out in the 10^{-9} sec range. Since this is much shorter than $\tau_{\rm op}$, it is unimportant here. Comparing the time scales, we get $\tau_J \ll \tau_{\rm op}$, meaning that the final connection of the superconducting regions into a coherent loop can be considered as instantaneous. Thus, this particular condition of the Kibble-Zurek mechanism is satisfied in our case, despite the relatively slow cooling rate of the experiment. It is the continuous presence of fluctuations, large and small, which occur at a rate much faster than $\tau_{\rm op}^{-1}$ throughout the experimental temperature range, which validates the analogy with the Kibble-Zurek scenario.

After the loop is closed, any further change of the flux trapped in the loop requires a global fluctuation of the whole loop, with an energy barrier $E_\pm = E_J + \phi_0^2/8L \pm n\phi_0^2/2L$, where the ($\pm$) depends on whether the flux increases or decreases [17]. Since L is large, $|\phi_0^2/8L \pm n\phi_0^2/2L| \ll E_J$, and the probability for a flux change continues to be dominated by the coupling energy. At $I_c \simeq 60\ \mu$A and $n = 30$, $E_\pm \simeq 30 k_B T$, enough to prevent flux jumps between metastable states of the whole loop. In the experiment, we see trapped flux up to $\sim 30\phi_0$. The energy needed to support $n\phi_0$ in the loop is $n^2\phi_0^2/2L \approx n^2 \times k_B T/10$. Putting $n = 30$ gives an energy of $90 k_B T$, which means that this large value of the flux would be unlikely to appear as a result of fluctuations at thermal equilibrium. It may be created only via a nonequilibrium mechanism, such as discussed above. The required energy comes from the coupling energy of the junctions during the closure of the loop (with our assumption $I_c\phi_0/\pi \sim 30 k_B T$, one needs only the energy of a few junctions).

Let us consider the magnitude of the spontaneous flux one can expect from the above mechanism. In the picture of the Kibble-Zurek mechanism, one usually considers the geodesic rule, which is just an assumption of minimal energy and, thus, a minimal phase gradient between adjacent segments [2,18]. In this model the average phase difference in a sense of a random walk is $\delta_{\rm rms} = \pi/2$, and the rms number of flux quanta in the loop is given by $n_{\rm rms} = \frac{1}{2\pi}\delta_{\rm rms} \times \sqrt{N}$ with N being the number of segments. In our experiment this should give $n_{\rm rms} \simeq 3.6$, however, we actually get $n_{\rm rms} = 7.4 \pm 0.7$. One possible reason is that the usual geodesic rule does not apply in the case of Josephson junctions, since the energy is proportional to $\cos(\delta)$ instead of the phase gradient squared. For example, in a junction, $\delta = \pi/2$ costs the same energy as $\delta = -3\pi/2$.

A somewhat better estimate of $n_{\rm rms}$ can be done using the probability distribution $P(\delta)$ of a junction. The average contribution of each junction to the total phase difference, in the sense of a random walk, is $\langle\delta^2\rangle^{1/2} = \{[\int \delta^2 P(\delta) d\delta]/[\int P(\delta) d\delta]\}^{1/2}$. In its coherent state, the maximal phase difference across the junction is $\leq 2\pi$ because otherwise a flux quantum is added or taken out of the loop. Consequently, the range of the integration is $(-2\pi, 2\pi)$. In order to take into account the spread of I_c's (and hence E_J's) between the junctions, we calculated $\langle \delta^2 \rangle^{1/2}$ for $6 < E_J/k_B T < 30$. A direct measurement of the spread of I_c's was impossible in our particular setup, however our experience with this type of junction suggests a spread by a factor of 3. Hence, taking a factor of 5 is on the safe side. The results for $\delta_{\mathrm{rms}} = \langle \delta^2 \rangle^{1/2}$ calculated this way are $1.31\pi < \delta_{\mathrm{rms}} < 1.37\pi$. We find that $9.6 < n_{\mathrm{rms}} < 10$, in reasonable agreement with the data. Thus, a scenario in which separate superconducting regions having different phases become connected preserving the total phase difference around the loop, seems a good starting point to understand the experimental results.

We are grateful to B. Ya. Shapiro and N. Gov for useful discussions, and to M. Ayalon and S. Hoida for technical assistance. This work was supported by the Israel Science Foundation, The Heinrich Hertz Minerva Center for HTSC, and by the VPR Technion Fund for Promotion of Research.

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