

## Search for spontaneous nucleation of magnetic flux during rapid cooling of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ films through $T_c$

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We describe an experimental search for spontaneous formation of flux lines during a rapid quench of thin  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  films through  $T_c$ . This effect is expected according to the Kibble-Zurek mechanism of a creation of topological defects of the order parameter during a symmetry-breaking phase transition. Spontaneously formed vortices were previously observed in superfluid  $^3\text{He}$ , while a similar experiment in superfluid  $^4\text{He}$  gave negative results. Using a high- $T_c$  superconducting quantum interference device, we measured both the magnetic flux in the sample during a quench with a sensitivity of  $20 \phi_0/\text{cm}^2$ , and the field noise which one would expect from flux lines pinned in the film. The sensitivity was sufficient to detect spontaneous flux at a level corresponding to  $10^{-3}$  of the prediction. Within our resolution, we saw no evidence for this effect.

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### I. INTRODUCTION

If a system undergoing a phase transition into an ordered state is quenched through the phase transition fast enough, topological defects can be created due to the evolution of uncorrelated regions of the newly formed phase, having different values of the order parameter. The defects appear at the boundary separating several coalesced regions of this kind. Such a scenario was proposed by Kibble<sup>1</sup> in the context of the grand unified theory, in order to describe the symmetry-breaking phase transition in the early universe. Zurek<sup>2</sup> developed this idea to predict the initial density of defects created during the phase transition and suggested specific experiments on condensed matter systems to test this scenario. Because of the generality of the theory of phase transitions, topological defect formation should occur in every physical system having a relevant symmetry breaking of the order parameter during the transition. The main arguments used by Zurek rely on the critical slowing down of fluctuations and the divergence of the coherence length near a second-order phase transition. Both quantities are influenced dynamically by the characteristic time required to complete the transition. In the last few years, experiments were carried out on several systems: first, nematic liquid crystals undergoing an isotropic-nematic transition<sup>4,5</sup> (in this case the topological defects are disclinations). Another system is liquid  $^4\text{He}$  crossing the  $\lambda$  transition as a result of rapid depressurization;<sup>6,7</sup> in this case the topological defects are quantized vortex lines. A third system is liquid  $^3\text{He}$  undergoing its superfluid phase transition.<sup>8,9</sup> In this experiment, the quantized vortices are formed during a thermal quench induced following an exothermic neutron-induced nuclear reaction. The experiments on liquid crystals and  $^3\text{He}$  gave results consistent with Zurek's prediction. In contrast, after early claims of observing this effect in liquid  $^4\text{He}$ ,<sup>6</sup> a recent improved experiment showed no spontaneous nucleation of vortices within the experimental resolution.<sup>7</sup> Here, we report an analogous experiment with a superconductor; the topological defects are quantized flux lines. Specifically, the ex-

periment aims to observe spontaneous flux lines generated during thermal quench of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) thin films through the normal to superconductor transition. The additional importance of this experiment is to test the "cosmological" scenario of Kibble and Zurek<sup>1,2</sup> in a system with a local gauge symmetry, where the theory is less clear than in a systems having a global gauge (e.g.,  $^4\text{He}$  and  $^3\text{He}$ ), due to the evolving gauge field ( $\mathbf{B}$ ) during the transition.<sup>10,11</sup> Within our resolution, we found no spontaneous vortex formation down to a level of  $10^{-3}$  of the predictions, which is in variance with the original theory of Zurek.

### II. THE KIBBLE-ZUREK MECHANISM IN A SUPERCONDUCTOR

Flux lines may become created spontaneously in type-II superconductors during a rapid quench through  $T_c$ . The most practical way is a thermal quench (which is more reliable from pressure quench in these materials, since the pressure dependence of  $T_c$  is rather weak). Low-temperature superconductors (LTS's) have a second-order phase transition which is well described by the Landau-Ginzburg theory and a rather small critical region, thus the anticipated initial flux-line density should be well predicted by Zurek's theory.<sup>2</sup> According to these predictions, the *initial* vortex density after the quench should be

$$n_i \approx \frac{1}{\xi_0^2} (\tau_0 / \tau_Q)^p. \quad (1)$$

Here,  $\xi_0$  and  $\tau_0$  are the coherence length and the relaxation time of the order parameter at  $T=0$ , respectively. The typical quench time  $\tau_Q$  is defined as  $\tau_Q = 1 / [(d\varepsilon/dt)_{\varepsilon=0}]$ ,  $\varepsilon$  being the reduced temperature. The exponent  $p$  is related to the critical exponents of the coherence length and the relaxation time as  $p = [2\nu / (\mu + 1)]$ , where  $\xi = \xi_0 |\varepsilon|^{-\nu}$ ,  $\tau = \tau_0 |\varepsilon|^{-\mu}$ . From an experimental point of view, the problem with LTS is that their coherence lengths are of the order of 100 nm and therefore the flux line density predicted by Eq.

(1) is quite small. A more favorable situation exists in high-temperature superconductors (HTS's) which have a much shorter coherence length ( $\xi_0 \approx 2$  nm). The phase transition of HTS is closer to the three-dimensional (3D)-XY model, and has a wider critical region than LTS, but as both the coherence length and the relaxation time diverge at  $T_c$ , that should be sufficient for spontaneous vortex formation (provided the quench is fast enough). Another important aspect unique to a superconductor is flux pinning, which can cause a significant reduction of the mutual annihilation of vortices and antivortices generated during the quench. Pinning can also prevent the flux from being expelled out of the film. Finally, within the last few years it was found that the order parameter in HTS has a predominant  $d$ -wave symmetry, while in metal LTS it is always  $s$ -wave. This type of pairing can lead to spontaneous flux generation in some special configurations (Ref. 13 and other references therein), but in a homogeneous material it should not affect the Kibble-Zurek scenario. The values pertinent to YBCO are  $\xi_0 \approx 1.6$  nm,  $\tau_0 \approx 5 \times 10^{-12}$  sec.<sup>14</sup> In our experimental setup, described below, we achieve a 20 K/sec cooling rate, giving  $\tau_Q \approx 5$  sec.

Because the phase transition in YBCO is not a pure Ginzburg-Landau type, closer to the 3D-XY model, the value of the exponent  $p$  is deduced from experiments that measured the exponents  $\nu$  and  $\mu$ .<sup>15-18</sup> We chose average values:  $\nu \approx 0.67$ ,  $\mu \approx 3.4$  (note that this is an unusual scaling), and then  $p \approx 0.3$ . Due to the short coherence length, the predicted initial flux-line density generated in the film by a thermal quench is very large:  $n_i \sim 10^{10}$  cm<sup>-2</sup> (vortices and antivortices). An important quantity is the reduced temperature  $\hat{\varepsilon}$  at which the system returns to the usual critical behavior during the quench, after an initial period during which it is out of equilibrium. Spontaneous flux nucleation takes place around this temperature, and thus determines the initial vortex density. According to the theory:  $\hat{\varepsilon} = (\tau_0 / \tau_Q)^{1/(\mu+1)}$ . In our experiment  $\hat{\varepsilon}$  is of the same order of magnitude as the width of the regime in which strong fluctuations can erase flux lines ( $\varepsilon_f \sim 10^{-3}$ ), as deduced from measurements of flux-line noise in thermal equilibrium.<sup>20</sup> However, as soon as the temperature decreases and the flux lines become pinned, one can hope to preserve a significant fraction of the initial flux-line density. In our system, the situation is more favorable in this respect than in superfluid <sup>4</sup>He, since there the regime of strong fluctuations is wider. Although the value of  $\hat{\varepsilon}$  is not necessarily determined by  $\varepsilon_G$  from the Ginzburg criterion<sup>21</sup> (in <sup>4</sup>He:  $\varepsilon_G \sim 0.2$ ) it is not reasonable that it can be smaller by more than a few orders of magnitude. In the recent experiment (Ref. 7)  $\hat{\varepsilon} \sim 10^{-5}$ . Moreover, there is no pinning of vortices in bulk <sup>4</sup>He which can help to preserve a significant fraction of these vortices. The freezeout coherence length  $\hat{\xi}$ , corresponding to  $\hat{\varepsilon}$ , is  $\hat{\xi} = \xi_0 (\tau_Q / \tau_0)^{\nu/(\mu+1)}$ . We get  $\hat{\xi} \sim 0.1$   $\mu$ m, of the same order of magnitude as the thickness of the films (50–300 nm). Therefore the initial vortex array is two-dimensional (but the physical system is 3D). In our experiment we can measure directly the difference between the number of vortices and antivortices, namely the net flux. If the simple picture of well defined phase regions with separation of  $\hat{\xi}$  and with a choice of a minimal phase gradient between these regions (the geodesic rule) being strictly cor-

rect, than the rms net flux should be  $\approx n_i^{1/4}$ . However, these arguments are unlikely to be strictly correct, especially in a real superconductor, due to the local gauge symmetry with the evolving magnetic field.<sup>10-12</sup> Moreover, the net flux cannot be determined from the simulations done until now<sup>22,23</sup> because these were performed using the simple geodesic rule and fixed periodic boundary conditions. For a plausible estimate of the net flux one must allow some relaxation of the geodesic rule.<sup>12</sup> This idea was proposed for first-order bubble collision, but it can be applied in principle to Zurek's theory at the stage at which uncorrelated phase regions with separation of  $\hat{\xi}$  are considered. If, for example, we relax the geodesic rule in such a way as to allow a variance of one random flux line in an area of  $100 \hat{\xi}^2$  (which may contain 100 vortices, antivortices, and homogeneous sites), the rms net flux will be determined through a random walk count of  $n_i/100$  flux lines. In this case, the net flux is  $\approx \frac{1}{10} \sqrt{n_i}$ . When relaxing the geodesic rule, we can count the net topological charge by integrating around the boundaries of the system, only after we identify the vortices inside, as otherwise it will not be self-consistent. Under this assumption, one sums the phase differences around any given loop while allowing, with some probability, a gradient of the phase between adjacent regions which is not minimal. The total topological charge will depend on this probability, and may acquire different values for the same initial arrangement of the regions having different values of the order parameter. One can see that even when  $n_i$  almost does not change (a small relaxation of the geodesic rule), the value of the net flux can become significant and rapidly approach its maximal statistical value of  $\sqrt{n_i}$ . Based on the above assumptions, we estimate the realistic order of magnitude of the net flux-line density as  $\frac{1}{10} \sqrt{n_i} = 10^4$  cm<sup>-2</sup>. Obviously, the net density of flux is not affected by mutual annihilation, since an equal number of vortices and antivortices disappear. It may decrease, however, as a result of flux lines being expelled out of the film. We show below that under the conditions of our experiment most of the net flux should remain in the film. Our setup, described below, can resolve net flux down to a limit of  $20 \phi_0/\text{cm}^2$ . Note that this sensitivity, with the assumption about the net flux being  $\frac{1}{10} \sqrt{n_i}$ , is sufficient to see the effect even if  $n_i$  is 4 orders of magnitude smaller than predicted by Eq. (1). The above estimation was done for a homogeneous phase transition. In our samples the superconducting transition is not completely homogeneous for two reasons: temperature gradients arising during the quench and a slightly different  $T_c$  in different regions of the film. We estimate the maximum temperature gradients as  $\nabla T \approx 1$  K/cm. The spread of  $T_c$  in the different regions of the film is of the same order. The homogeneous approximation holds if  $v_T > \hat{s}$ ,<sup>3,24</sup> where  $v_T$  is the velocity of the phase transition front propagating across the film and  $\hat{s}$  is the characteristic speed at which superconducting order-parameter fluctuations propagate at  $\hat{\varepsilon}$ . This condition is identical to imposing the demand that  $\nabla T < T_c \hat{\varepsilon} / \hat{\xi}$ . The estimated value of the right-hand side of the inequality is more than  $10^3$  K/cm, and we therefore conclude that the homogeneous approximation is correct.

One more possible experimental geometry is a superconducting loop. One can try to measure spontaneous flux gen-

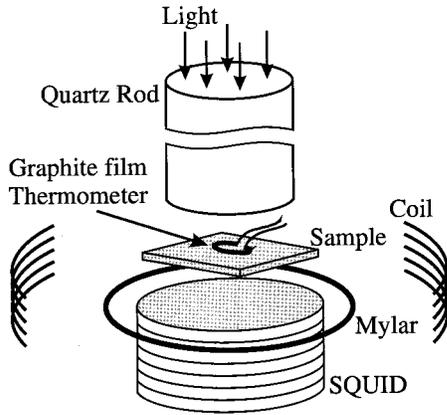


FIG. 1. A schematic layout of the essential part of the experimental system. For clarity, only the edge of the mylar barrier is shown. The sample itself is held inside a light-tight plastic holder to prevent the light from reaching the SQUID. The assembly shown is surrounded by several  $\mu$ -metal shields.

erated during a rapid thermal quench of this loop. The basic idea here is the same, namely that uncorrelated regions will be generated during the quench, with random phases of the order parameter in each one of them. The accumulated phase difference around the loop will create a supercurrent and magnetic flux through the center of the loop. If the width  $d$  of superconductor forming the loop is of the order of magnitude of  $\xi$ , the average (rms) number of flux quanta generated in the loop is  $n_\phi = 1/4 \sqrt{L/\xi}$ , where  $L$  is the circumference of the loop.<sup>3</sup> In our typical pattern,  $L \approx 20$  mm, and  $d \approx 10$   $\mu$ m (somewhat bigger than  $\xi$ ) so we can substitute  $d$  instead of  $\xi$  and get  $n_\phi \approx 11$ . This simple argument is subject to the same uncertainties as the ones encountered above for bulk film, due to the presence of the magnetic field.

### III. EXPERIMENTAL SETUP

In order to measure the flux, we used a high- $T_c$  superconducting quantum interference device (SQUID) placed close to the superconducting film. The SQUID can detect (a) the net-flux nucleated as the film is quenched through  $T_c$ , and (b) the field noise caused by random hopping of these flux lines. The sign of the net flux should be random for each individual quench. In contrast, the rms power density of the noise spectrum is the same for vortices and antivortices, and should characterize the *total* density of flux, rather than the net density.

The essential part of the experimental setup is shown in Fig. 1. It is basically a cryostat divided into two cells separated by a thin mylar barrier. The SQUID and the sample are mounted on the two sides of the barrier, facing one another. The distance between the SQUID and the superconducting film is about 1 mm. The cryostat is immersed in liquid nitrogen at 77 K. The whole system is carefully shielded from the earth's magnetic field by several  $\mu$ -metal layers arranged inside a soft iron container. The residual magnetic field in the cryostat was less than 0.2 mG. Additional small coil adjacent to the sample was used to null this field, as well as for testing the field dependence of the results. The reason for using two separate chambers is that in this arrangement the SQUID

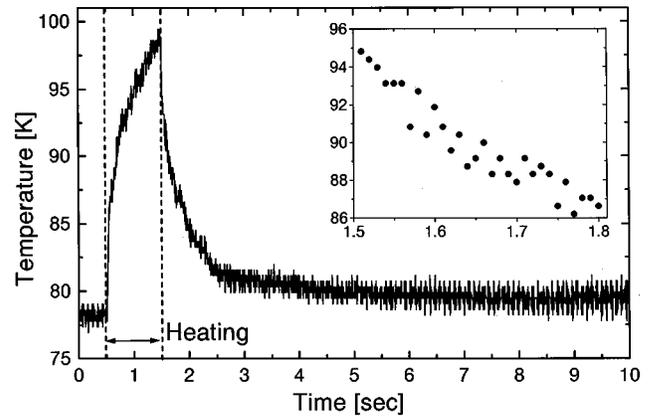


FIG. 2. A typical profile of the temperature vs time during a heating-cooling cycle. The inset shows an expanded view of the cooling profile near  $T_c$ .

remains at a constant temperature, while the sample can be heated and cooled independently. To avoid spurious magnetic fields generated by the current used in resistive heating, the film is heated using a focused light beam. The light is introduced into the cryostat via a quartz rod, terminating at about 2 mm from the sample. The light illuminates the whole sample area ( $10 \times 10$  mm<sup>2</sup>) and is confined within the plastic holder tube of the sample. In order to achieve maximum efficiency of the heating, the superconducting film is coated by a graphite layer which absorbs the light.

Cooling of the sample after the heating stops is via a strong thermal link to liquid nitrogen, through helium exchange gas present in the cell. The cooling rate through  $T_c$  can be regulated by changing the pressure of the gas, and hence its thermal conductivity. It takes about 1–2 sec to heat the sample above  $T_c$  ( $\approx 90$  K). Cooling begins immediately when the light is turned off (by a shutter at the top of the cryostat). The maximum cooling rate through  $T_c$  is 20 K/sec. Because the heating and cooling is done mainly perpendicular to the plane of the substrate, the temperature of the sample is approximately the same along its lateral dimensions. Moreover, the critical slowing down of the fluctuation in the film near  $T_c$  does not affect the cooling rate. It is possible to keep the SQUID's side of the cell in vacuum to avoid any heat leak from the sample to the SQUID, in practice, it was not necessary.

To measure the temperature of the sample in real time during the heating-cooling cycle we used a thin graphite strip painted on the back side of the substrate. Its heat capacity is small, similar to that of the superconducting film, and the changes of its resistance with temperature enable us to measure the true temperature of the film in real time. The temperature dependence of the resistance of the graphite was calibrated against a diode thermometer. A typical temperature measurement during the quench is shown in Fig. 2. During the heating period, the graphite strip may be hotter than the film by about 1 K, because it faces the light directly, but the cooling rate is approximately the same. We usually raised the temperature of the graphite to about 100 K to ensure that all the film was heated above  $T_c$ . We ascertained that the small measuring current through the graphite strip did not affect the output of the SQUID, which was the same

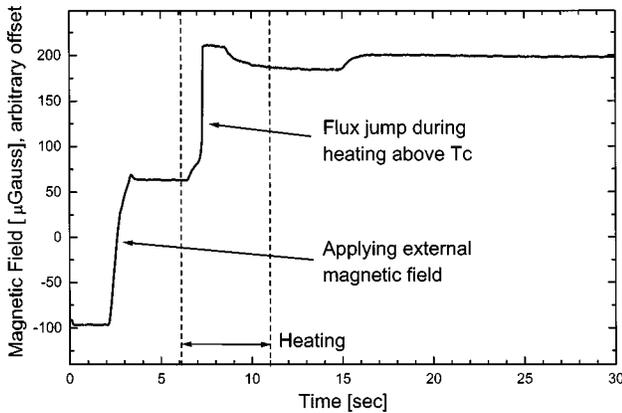


FIG. 3. Response of the system to an application of an external field of 0.3 mG. Initially, the film is at 77 K, and after the heating-cooling cycle it cools back to this temperature.

with or without temperature measurement.

At different times through the experiment, we used two kinds of HTS dc SQUID's: a commercial M-2700 unit made by Conductus, and a grain-boundary SQUID made in our lab. Both were operated in a flux locked loop (FLL). The commercial SQUID has a magnetic field sensitivity of  $2 \times 10^{-4}$  Gauss/ $\phi_0$  and an integrated pickup loop with an area close to 1 cm<sup>2</sup>. The homemade SQUID, made from a thin YBCO film on a SrTiO<sub>3</sub> bicrystal substrate, has a similar field sensitivity, however, its noise is larger by a factor of 30 than the commercial SQUID. The advantage of using the homemade SQUID is that it can be mounted closer to the sample, as it is not encapsulated. The distance between the homemade SQUID and the YBCO film is about 0.5 mm, while for the commercial unit it is 1 mm. Except for the aforementioned signal-to-noise (S/N) ratio, the results presented below were the same with both SQUIDs.

Working with a HTS SQUID enabled us to keep the whole experiment at 77 K and to perform the fast thermal quench as described. Measurement of net flux was done continuously during the heating and cooling cycle, but the noise becomes well defined just close to equilibrium (i.e., at the end of the cooling). If a net flux is generated in the film during the quench it should be seen as an offset relative to the zero flux state prevailing when the film is above  $T_c$ . As stated above, according to the Zurek scenario this flux should increase in amplitude with the cooling rate while its sign should be random from one quench to the next. If a large number of vortices and antivortices is preserved in the film at the end of the cooldown to 77 K, the residual flux noise should be stronger compared to a situation when the film is relatively clean from vortices (for example, after a very slow cooling or when the temperature is above  $T_c$ ).

We now turn to estimate the sensitivity of the experiment. The coupling ratio between the pickup loop and the SQUID is  $10^3$  and the dc-flux level change which one can clearly resolve is  $\approx \phi_0/100$ . According to our measurements of the screening of the SQUID by the sample, a single vortex in the sample induces (on average) about  $0.5\phi_0$  into the pickup loop. Dividing by the coupling ratio we get a net flux resolution (in the sample) of  $\sim 20$  net  $\phi_0/\text{cm}^2$ . This measured value of the coupling coefficient is consistent with calculations.<sup>19,20</sup> In Fig. 3, we show an example of a se-

quence in which an external field was applied while the sample was cold. The field change picked up by the SQUID is about half of that applied, which serves to estimate the coupling coefficient. As the film is heated above  $T_c$ , the whole field penetrates the sample and is picked up by the SQUID. We found the same coupling coefficient by following the reverse procedure, namely cooling the film in a field, then turning off the external magnetic field, and finally releasing the remanent flux pinned in the sample by heating it above  $T_c$ . From these we conclude that the samples have strong flux pinning and that the coupling coefficient of flux into the SQUID is about 0.5. We used the same method to test the superconducting continuity of the ring patterns. In addition, we found that external magnetic fields of up to 10 mG become trapped during cooldown in the film very close to  $T_c$ . Thus any spontaneously generated flux remaining in the film after it is cooled to a temperature outside the interval  $\hat{\epsilon}$  below  $T_c$ , should survive the cooldown to 77 K. A net flux-line density of  $10^4 \phi_0/\text{cm}^2$  is equivalent to a field of 2 mG. The pinning site density in similar films was estimated in Ref. 20 (and in references therein) as  $1-6 \times 10^{10} \text{ cm}^{-2}$ . Thus we conclude that most of the net flux generated during a quench should remain pinned in the film during the cooldown.

To estimate the expected field noise in the film, we rely on noise measurements in YBCO thin films, done with a SQUID having field sensitivity similar to ours.<sup>20</sup> It was found that the noise power spectrum is linearly proportional to the magnetic field in the film and has a  $1/f$  dependence. The magnitude of the rms field noise (at 10 Hz) with 10 G ( $10^8$  vortices/cm<sup>2</sup>) was  $10^{-8}-10^{-7} \text{ G}/\sqrt{\text{Hz}}$  at 77 K (the exact number depending on the sample). The field noise of our SQUID is  $\approx 3 \times 10^{-9} \text{ G}/\sqrt{\text{Hz}}$ . Thus we expect to resolve a contribution to the noise in the  $1/f$  regime for a total flux-line density equal to or larger than  $10^6 \text{ cm}^{-2}$ . This would be possible if  $10^{-4}$  (or more) of the initial density survives. The estimates done so far refer to unpatterned films ( $1 \times 1 \text{ cm}^2$  area). The limit of sensitivity in experiments on films patterned into loops (with diameter of  $\approx 7 \text{ mm}$ ) is  $\sim 10\phi_0$  net.

Finally, the residual magnetic field in the experimental cell (before nulling), was found to be less than 0.2 mG. Since any nulling procedure is imperfect, a small constant field is always present in the film when cooling through  $T_c$ . In order to evaluate the effect of any residual external field on our data, quenches were performed under different fields in the mG range, varying systematically both in magnitude and in the sign.

#### IV. RESULTS AND DISCUSSION

In this study, we used several types of *c*-axis oriented epitaxial YBCO films: (1) dc-sputtered YBCO on (100) SrTiO<sub>3</sub>; (2) DC-sputtered YBCO on (100) MgO; (3) laser ablation deposited YBCO on (100) SrTiO<sub>3</sub>; and (4) dc-sputtered YBCO on (001) NdGaO<sub>3</sub>. We have also tested a sintered YBCO ceramic sample. There are several important differences between the various types of films. Generally, sputtered films have better crystallinity than the ones grown by laser ablation. The degree of crystallinity is a measure of the defect density in the film, and thus the density of the

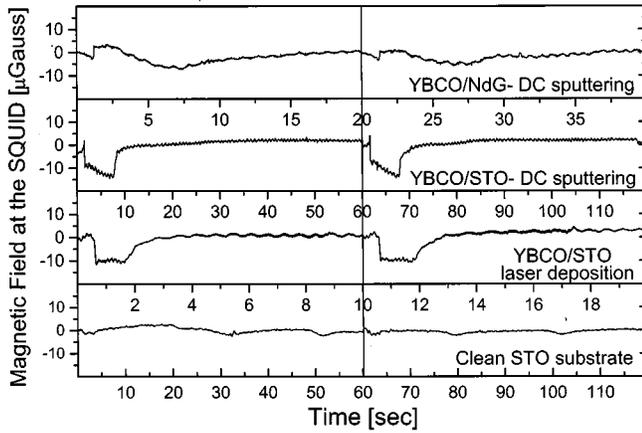


FIG. 4. Typical time dependence of the flux picked up by the SQUID for several samples undergoing heating-cooling cycles. The vertical line in the middle of the figure separates two consecutive heating-cooling cycles. The heating interval takes 1–2 sec at the beginning of each cycle. Note that the cooling rates for each sample are different, as shown by the different times on the respective horizontal axes.

pinning sites. Films grown on (001)  $\text{NdGaO}_3$  are unidirectionally twinned, while those grown on other substrates are bidirectionally twinned. In addition, the films on (001)  $\text{NdGaO}_3$  are lined by unidirectional nanocracks, perpendicular to the twins, and can be thought of as composed of superconducting strips several  $\mu\text{m}$  wide, separated by the nanocracks.<sup>25</sup> Consequently, in those films all the flux lines are very close to some boundary and can be therefore expelled from the film during cooldown. Films of each type, and of varying thickness in the range 50–300 nm, were tested both as grown ( $10 \times 10 \text{ mm}^2$ ) and after patterning. The patterns used were: (1) discs (8-mm diameter); (2) rings of 7-mm diameter and 10–100- $\mu\text{m}$  width; (3) an array of 100 rings of 200- $\mu\text{m}$  diameter and 3- $\mu\text{m}$  width.

An experiment is taken by performing a thermal quench as described while recording the SQUID's FLL voltage output vs time, or equivalently, vs temperature. For each sample we recorded many consecutive quenches. Typical examples of such measurements performed on three different kinds of samples are shown in Fig. 4, along with a reference measurement, that of a substrate coated with graphite, but without any superconducting film. We show here two consecutive quenches for each sample. It is clearly seen that there are small flux jumps during the heating and cooling, but no residual flux after the samples are cooled. These flux jumps may be different from sample to sample. However, for any given sample the flux jumps seen here appear the same through all the quenches, both in the sign and magnitude. Moreover, these flux jumps are not affected by external magnetic fields or from the field present at the feedback-modulation coil of the SQUID. Thus these jumps are definitely not related to the Kibble-Zurek mechanism. Possible origin of this effect, which occurs near  $T_c$ , will be discussed elsewhere. The fact that we see no residual flux at the end of the cooling shows that within our resolution no flux lines were spontaneously nucleated in the film during the quench. Our estimate of the predicted net spontaneous flux-line density is  $\sim 10^4 \text{ cm}^{-2}$ . This should give a magnetic field of  $\sim 2$

mG at the sample (1 mG at the SQUID, which is about  $5 \times 10^2$  times bigger than our resolution). If spontaneous flux of this magnitude was created, we should have seen a large signal during the cooling (about a factor of 100 larger than the reproducible flux jumps shown in Fig. 4). Even if the annihilation of vortices and antivortices and the expelling of flux out of the film is very fast, we should have still seen a transient signal of varying sign and magnitude as the film cooled through  $T_c$ . Such signals were not observed even at the fastest recording rate ( $10^4$  data points/sec, corresponding to a temperature change of  $2 \times 10^{-5}$  in  $\varepsilon$  between successive points) in the vicinity of  $T_c$ . At this sampling rate, the time  $t_m$  between successive data points is 2 orders of magnitude smaller than the time  $\hat{t} = (\tau_0 \tau_Q^\mu)^{1/(\mu+1)}$  during which the system cools from  $T_c$  to  $\hat{\varepsilon}$  during the quench. In our system,  $\hat{t} \approx 10^{-2}$  sec (the recording rate was limited by the minimal integration time constant of the flux locked loop). Similarly, negative results were found also with the films patterned into rings. Finally, we did not observe any increase in the magnitude of the noise at the end of the cooldown following a quench.

In conclusion, in our experiments we found no evidence for spontaneous flux-line formation down to a limit of, at least,  $10^{-3}$  of the predicted initial density. One possible reason for the negative result is that the relation for  $n_i$  [Eq. (1)] is just a first estimation and in practice it may be smaller. For example, results obtained in experiments on  $^3\text{He}$  and  $^4\text{He}$  and also in numerical simulations are smaller by as much as 1 or 2 orders of magnitude than predicted by Eq. (1). The negative results in  $^4\text{He}$  may be a consequence of a fast decay of vortex loops which takes place before the measurement even begins. This scenario follows from the calculations done by Williams.<sup>26</sup> As he pointed out, a similar mechanism may also cause a decay of vortex loops in a superconductor. In our case, vortex loop can be thought of as a vortex-antivortex pair. Since this scenario involves only loops, no initial imbalance between the number of vortices and antivortices is permitted. If, however, vortices and antivortices are not created in equal numbers (as loops), then the imbalance between the number of vortices and antivortices (the net flux) cannot decay via this mechanism. There is no restriction within the Kibble-Zurek picture that the number of vortices and antivortices should be equal. In our experiment, the mechanism proposed by Williams would be reflected in the decrease of the total vortex density and hence in the residual noise level at the end of the quench, but it will not affect the anticipated *net* flux density (our main measuring technique). The theory of Zurek implies that after the initial creation, the density of vortices will decay as  $\sim n_i \hat{t}/t$ , where  $t=0$  at  $T_c$ . In the  $^4\text{He}$  experiment, the measurement began at a time which is much bigger than  $\hat{t}$ , while in our fast measurements we took data with a time resolution  $t_m \ll \hat{t}$ , and we covered the whole interval from  $t < 0$  up to  $t \gg \hat{t}$ . Thus we should have seen something at least during the initial stage, disrespectful of the details of any subsequent decay. In addition to the work of Williams, a reexamination of the Zurek scenario for the  $^4\text{He}$  case was done by Karra and Rivers.<sup>27</sup> Their calculation yields a vortex density consistent with the negative experimental result. It is not clear what should be the

result of a similar calculation for a 3D high- $T_c$  superconductor. A very interesting simulation of a rapid cooling of a normal spot created inside a 2D superconductor (including pinning) was carried out recently.<sup>28</sup> In particular, this simulation shows the great importance of pinning in preserving a significant fraction of the vortex population originating in the quench, essentially *ad infinitum*. It would be highly desirable to extend this work to the case where the topological charge during the quench is not conserved. Finally, we point out that the very question of two separate superconductors having a

well defined phase difference before coming into contact is still under debate.<sup>29</sup> We hope that our results may shed some light on this interesting issue.

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