Observation of Magnetic Flux Generated Spontaneously During a Rapid Quench of Superconducting Films

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We report observations of spontaneous formation of magnetic flux lines during a rapid quench of $YBa_2Cu_3O_{7-\delta}$ films through T_c. This effect is predicted according to the Kibble-Zurek mechanism of creation of topological defects of the order parameter during a symmetry-breaking phase transition. Our previous experiment, at a quench rate of 20 K/s, gave null results. In the present experiment, the quench rate was increased to $> 10^8$ K/s. The amount of spontaneous flux increases weakly with the cooling rate.

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A certain class of grand unified theories describes the early Universe in terms of a series of symmetry-breaking phase transitions. In that context, Kibble [1] predicted that if a system having a complex order parameter is quenched through a phase transition into an ordered state, topological defects will be created. This is due to the evolution of uncorrelated regions of the newly formed phase, each region having different values of the order parameter. The defects appear in between several coalesced regions of this kind. Zurek [2] developed this idea to predict the initial density of defects as a function of quench rate and suggested specific experiments on condensed matter systems to test this scenario. The natural candidates for such tests are superfluids and superconductors, in which the topological defects are quantized vortex lines. Other related systems are liquid crystals undergoing an isotropic-nematic transition [3,4], where the defects are disclinations. Superconductors have an added degree of complexity, due to the presence of the gauge field **A** which evolves with time. Results of various experiments done so far are not unambiguous; spontaneously generated vortices were observed in superfluid ^{3}He [5,6] but not in ^{4}He [7]. Experiments with homogeneous superconductors have so far shown null results [8]. Experiments done with superconducting rings [9] were done in a regime where usual thermal fluctuations dominate, rather than Zurek's mechanism. Experiments using Josephson junctions [10,11] gave results broadly consistent with the Zurek scenario. However, these are intrinsically inhomogeneous systems which do not fall into the class of systems directly comparable with this theory. Here, we report the results of a new, improved experiment with superconducting films [8].

According to the prediction, the total amount of spontaneous flux is proportional to $1/\xi_0^2$, where ξ_0 is the coherence length. It is therefore advantageous to use high temperature superconductors which have a much smaller ξ_0 than conventional superconductors. We therefore used epitaxial c-axis oriented $YBa_2Cu_3O_7$ films with $T_c \simeq 90$ K. The films, typically 300 nm thick, were grown on a $SrTiO_3$ substrate and patterned into a disk, 6 mm in diameter. Another advantage of using epitaxial films is that they contain no large angle grain boundaries, thus excluding any flux generation associated with defects [12]. The basic experimental setup is described in Ref. [8]. Briefly, the sample is placed atop the sensing coil of a high temperature superconductor (HTSC) SQUID magnetometer, at a distance of 1 mm. In our arrangement the SQUID remains at a temperature of 77 K, and is not affected by the temperature of the sample which can be heated and cooled independently. To avoid spurious magnetic fields generated by electrical current used in resistive heating, the film is heated above T_c using a light source and cools by exchanging heat with its environment. The system is carefully shielded from the Earth's magnetic field, with a residual magnetic field of less than 0.05 mG. An additional small coil adjacent to the sample was used to test the field dependence of the results. Instead of $\sim$1 s long illumination from a quartz lamp used to heat the sample in our previous work [8], the light source in the present experiment is a pulsed Nd:YAG laser [13]. Single pulses, 10^{-8} s long, were used to heat the film. After passing through a diffuser, the laser pulse passes through the substrate and illuminates homogeneously a 9 mm diameter area of the film, larger than our sample. At a laser wavelength of 1.06 μm, the $SrTiO_3$ substrate is transparent and practically all the light is absorbed in the film. Hence, only the film heats up, while the substrate remains near the base temperature of 77 K. The 1 mm thick substrate has a heat capacity about 10^3 larger than that of the film. Therefore, an energy of $\sim$mJ is sufficient to heat the film above T_c, rather than a $\sim$J used previously [8]. The heat from the film escapes into the substrate, which acts as a heat sink. This strongly reduced thermal mass which is cooled allows us to achieve cooling rates in excess of 10^8 K/s, 7 orders of magnitude faster than previously [8]. The cooling rate at T_c can be varied by changing the amount of energy delivered by the laser pulse (see Fig. 1).

As Fig. 1 shows, increasing this energy reduces the cooling rate. The cooling rate was determined by monitoring the time dependence of the resistance of a reference sample following a laser pulse. The reference sample is a similar film of underdoped $YBa_2Cu_3O_y$, having T_c of 60 K, which is not superconducting in our temperature range. Around 90 K, this sample has thermal properties [14,15] similar ($\pm$ 10%) to those of the films used in the experiment. This uncertainty sets the lower limit on the error bars of the cooling rate. Because heat flow into the substrate takes place in a direction normal to the plane of the film, the temperature of the sample is approximately the same along its lateral dimensions.

Achieving as high a cooling rate as possible is extremely important in order to enter the regime where the Zurek scenario applies. To observe the effect, the system needs to be out of equilibrium over a temperature interval wider than the critical regime near T_c. Because of the anisotropy of the superconducting properties of YBCO, our films are effectively 2D near T_c. We therefore expect that quantized vortices will develop only perpendicular to the film surface. The 2D Ginzburg-Landau (GL) model yields a 10^{-2} K as the width of the critical regime [16] (the 2D XY model gives an even smaller value). At quench rates between 10^7 and 10^8 K/s the system remains out of equilibrium over an interval of 0.1–0.2 K of T_c. Thus, the conditions for observing this effect are satisfied in our case. Using the small coherence length of HTSC, the predicted initial flux line density n_i generated in the film by a thermal quench is very large according to this scenario; $n_i = 10^{11}$–10^{12} cm^{-2} . This includes both vortices and antivortices, with the lower value corresponding to the GL model, and the larger to the 2D XY model. In our experiment we measure directly the difference between the number of vortices and antivortices, namely, the net flux. If the picture of regions having a well-defined phase, and with the choice of a minimal phase gradient between the regions (the geodesic rule [17]) is correct, then the net rms flux should scale as $n_{\rm net} \sim (L)^{1/2}(n_i)^{1/4}$, where L is the length of the sample perimeter. In terms of the quench rate dT/dt, $n_{\rm net} \sim (dT/dt)^{1/8}$. We point out that this weak dependence leads to a predicted $n_{\rm net}$ which increases only by 33% while the quench rate increases by an order of magnitude. In the range of our experiment, the net flux density is predicted to be $\sim 10^2$ ϕ_0/cm^2. The intrinsic noise level (integrated over the bandwidth) of our magnetometer is equivalent to a flux noise of $\sim$5 net ϕ_0 (see Fig. 3). Thus, the effect should be observable. It should be noted that our measuring system can detect only the net flux "frozen" in the film, due to the fact that the film's total cooling time is of the order of 1 μs, while the SQUID system responds on a time scale of about 10 μs. Flux will be frozen in the film if the pinning site density is much larger than the flux density. The pinning site density in similar films was estimated in Ref. [18] (and in references therein) as (1–6) $\times$ 10^{10} cm^{-2} $\gg n_{\rm net}$. Since pinning in YBCO films is very strong at temperatures below the critical regime, we conclude that the net flux generated during the quench should remain inside the film.

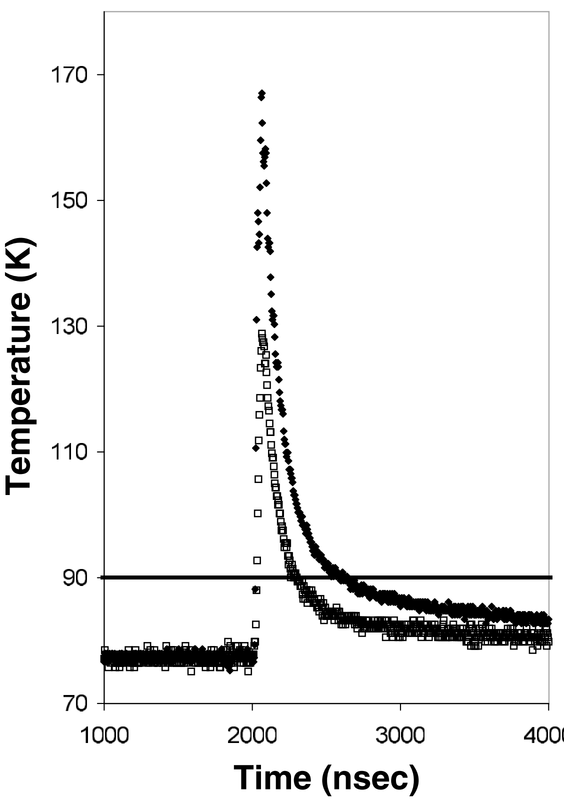

FIG. 1. Temperature cycle of the film after a laser pulse. The energy of the pulse is 7.1 mJ (solid symbols) and 3.1 mJ (open symbols). The horizontal line is T_c for our sample. Note that the cooling rate through T_c is slower for the high energy pulse.

In a typical experiment, the SQUID's output is recorded versus time as the sample cools following a thermal quench. Such measurements were performed both on superconducting samples and on a control sample. Indeed, net flux was observed with the superconducting film while no flux was seen in the control experiment. Figure 2 shows raw data from such measurements, taken at a quench rate of 10^8 K/s. According to the Zurek scenario, the sign of the net flux should be random from one quench to the next. Figure 2 clearly shows that this indeed is the case. Each of the raw data points in Fig. 2 taken with the film includes a contribution due to the appearance of spontaneous flux, mixed with the noise. To deconvolute these two contributions, we used the measured probability distribution of the noise $N(z)$ (see Fig. 3) and the probability distribution of the measured raw data, $R(z)$. Here, z is the output voltage of the magnetometer. The probability distribution function of the net spontaneous flux, $F(z)$, was extracted using the relation $F(z) = \int R(z + y)N(y)dy$. To convert z to absolute flux units, we used the measured sample to SQUID flux

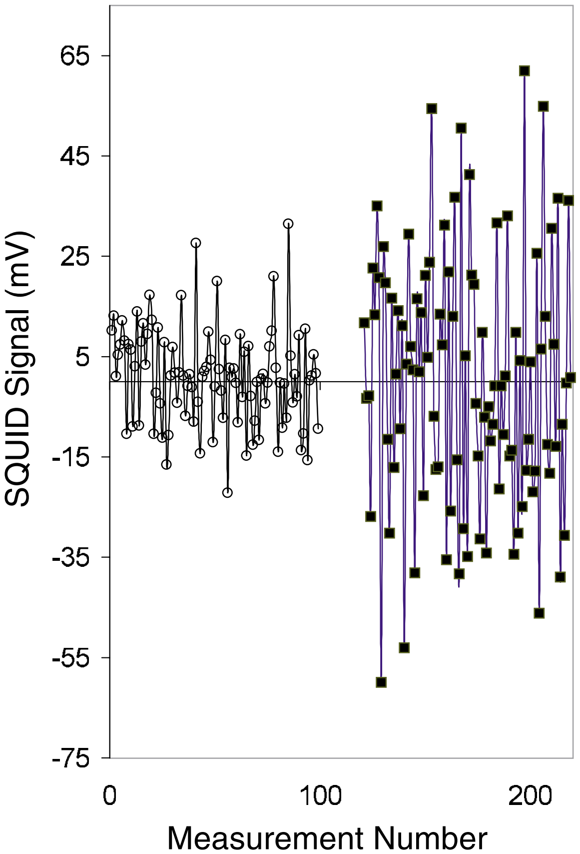

FIG. 2 (color online). Typical sequence of 100 raw data readings of the magnetometer, each following a separate quench at a rate of 10^8 K/s. Open symbols: control sample (“noise”); solid symbols: superconducting film. The conversion into flux units is 4 mV$/\phi_0$ for the noise and 1 mV$/\phi_0$ for the signal from the superconducting film. The lines connect successive data points.

coupling factor of 1 mV$/\phi_0$. This factor is different for the signal (1 mV$/\phi_0$) and noise (4 mV$/\phi_0$), since only $\sim$0.25 of the flux in the sample couples to the magnetometer, while all the measured noise originates in the magnetometer itself. Hence, the conversion to absolute flux units must be carried out only after the deconvolution. Because of these different weights, the true S/N in Fig. 2 is actually much better than it looks. This can be seen in Fig. 3, where a typical distribution of spontaneous flux (after deconvolution) is shown, as well as that of the noise. It can be seen that the distribution of the signal is symmetric about zero flux, as expected from this scenario. In order to check for the effect of any residual field, our measurements were repeated under different fields up to 10 mG, about 10^3 times larger than our residual field. The inset of Fig. 3 shows that the magnetic field has no significant influence on our results.

The net flux distribution width versus the cooling rate is shown in Fig. 4. The magnitude of spontaneous flux seems to increase weakly with the cooling rate. The solid line shows the $(dT/dt)^{1/8}$ dependence predicted by Zurek

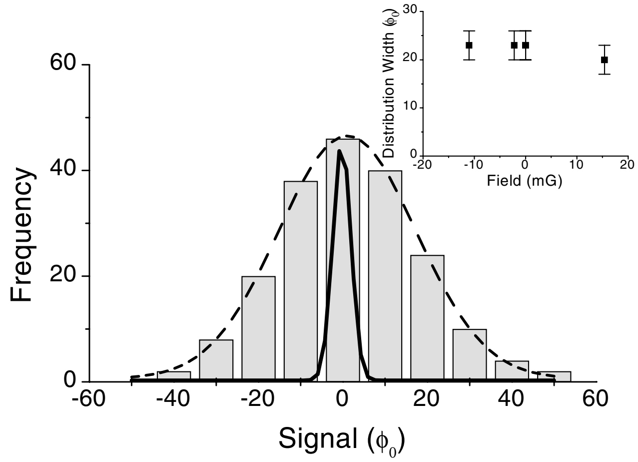

FIG. 3. Typical histogram of spontaneous flux from several hundred quenches. The dashed line is a Gaussian fit. The solid line is a fit to the noise measurements taken at the same conditions. The inset shows that the distribution width does not depend on the external field. The error bars in the inset are the uncertainty of the fits. The cooling rate for all the data in the figure (including the inset) is $\simeq 10^8$ K/s.

[2,19], with a prefactor given by Rudaz *et al.* [17]. To produce this curve, the theoretical prediction was scaled down by a factor of 8. The fact that the measured net flux is very small, $\sim 10^{-10}$ of the predicted total flux, further implies that the geodesic rule is valid in a nonequilibrium regime. The validity of this rule was not considered obvious [20]. Relaxing this rule sharply increases the predicted amount of net flux, which was not observed [8]. For completeness, we mention that another scenario [21] exists, predicting an amount of spontaneous flux which is much smaller than that given by [2,19]. Hence, this scenario is below our present limit of detection. Extrapolating our data shown in Fig. 4 down to a cooling rate of 20 K/s, that of Ref. [8], gives a predicted flux density of $\sim 3\ \phi_0/\mathrm{cm}^2$ for that experiment. This value is

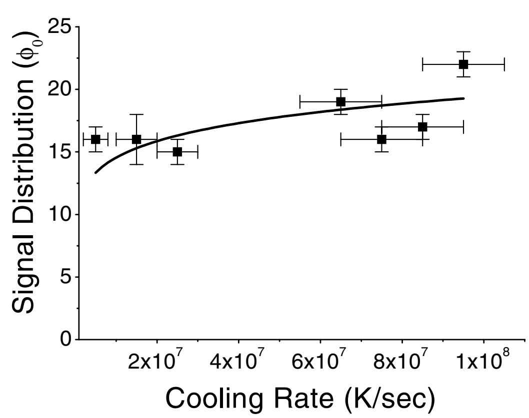

FIG. 4. Dependence of the distribution of spontaneous flux versus the cooling rate. The vertical error bars are those of the fit to a Gaussian distribution. The solid line is the prediction of Refs. [2,17] scaled to fit the data.

very close to the noise level, and thus explains the null result obtained in our previous work [8]. Finally, we carried out several checks to see how the magnitude of a signal is influenced by temperature gradients. The presence of temperature gradients is important with respect to the homogeneous approximation [19,22]. We estimate our maximum temperature gradient as $\nabla T \sim 1$ K/cm parallel to the film surface, similar to the spread of T_c across the film. Under these conditions, the homogeneous approximation is valid in our experiment. In these additional experiments, we created intentional temperature gradients in order to check whether the scaling factor between theory and experiment cited here is a result of such gradients present in our film. We found that this was not the case [23].

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