Observation of a New Mechanism of Spontaneous Generation of Magnetic Flux in a Superconductor

Ariel Maniv,* Emil Polturak, Gad Koren, and Yuri Bliokh
Physics Department, Technion - Israel Institute of Technology, Haifa 32000, Israel

Björn Biehler, Bernd-Uwe Runge, and Paul Leiderer
Physics Department, Konstanz University, 78457 Konstanz, Germany

Boris Shapiro and Irena Shapiro
Physics Department, Bar-Ilan University, Ramat-Gan 52900, Israel
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We report the discovery of a new mechanism of spontaneous generation of a magnetic flux in a superconductor cooled through T_c. Values of the spontaneous flux appear random from one cooldown to the next, following a Gaussian distribution. The width of the distribution increases with the size of the temperature gradient in the sample. Our observations appear inconsistent with the well-known mechanisms of flux generation. The dependence on the temperature gradient suggests that the flux may be generated through an instability of the thermoelectric superconducting-normal quasiparticle counterflow.

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With the exception of ferromagnets, a spontaneous appearance of a magnetic field in a physical system is a highly unexpected phenomenon. Yet, such a phenomenon was observed in superconductors cooled through T_c [1–6]. In one case, the spontaneous magnetic field appeared as a consequence of the *d*-wave symmetry of the order parameter of high temperature superconductors (HTSC) [1,4]. In another case, the spontaneous field was generated by cooling the superconductor through T_c under non equilibrium thermal conditions [2,3,5,6]. Here, we report a new, unexpected appearance of a spontaneous field, which occurs in a superconductor cooled in the presence of a thermal gradient.

Although the effect described below appears completely unrelated, the original motivation for this experiment followed our previous work on the Kibble-Zurek cosmological scenario [7,8]. One of the key assumptions of this scenario is that the temperature within the sample is uniform. The limit on the size of ∇T, the temperature gradient across the sample, set by Kibble and Volovik [9], is that $\nabla T < T_c\hat{\varepsilon}/\hat{\xi}$. Here T_c is the transition temperature, $\hat{\varepsilon} = \frac{\hat{T}-T_c}{T_c}$ and $\hat{\xi}$ are the reduced temperature and coherence length, respectively, at the temperature $\hat{T}$ at which fluctuations of the order parameter return to thermal equilibrium [8]. Our experiment was designed to see what happens to the formation of topological defects once this criterion is not satisfied.

The experimental setup is the same as described in Ref. [4], with the exception of a nonuniform heating, generating intentional temperature gradients in the sample. Briefly, our samples were 300 nm thick *c*-axis oriented $YBa_2Cu_3O_7$ films with $T_c \simeq 90$ K, grown on a $SrTiO_3$ substrate. The samples were placed atop the sensing coil of a HTSC SQUID magnetometer. 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. The film is heated above T_c using a light source and cools by exchanging heat with its environment. The light source is a pulsed YAG laser [10]. Single pulses (FWHM $\sim$ 10 ns) were used to heat the film. The laser pulse passes through the substrate and illuminates *nonuniformly* a selected area of the film. 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. The heat from the film escapes into the substrate, which acts as a heat sink. This small thermal mass of the film allows us to achieve cooling rates in excess of 10^8 K/sec. The cooling rate at T_c can be varied by changing the amount of energy delivered by the laser pulse. The system is carefully shielded from the earth's magnetic field, with a residual field of less than 50 μG. An additional small coil adjacent to the sample was used to test the field dependence of the results, at fields ranging from less than 50 μG up to 60 mG.

Nonuniform illumination was generated either by using a nonuniform light beam, or by covering some part of the sample. An example of one such arrangement is shown in the inset of Fig. 2. Here, the strongly illuminated area is a stripe across the film. In another configuration, the perimeter of the film was masked, while an area of 4 mm in diameter in the center was exposed to the beam. Qualitatively, the results presented here do not depend on the exact illumination profile. Under such nonuniform illumination, the film cools down in a two stage process. As measured previously [3], in the first stage the heat deposited by the laser pulse in the film is dumped into the $SrTiO_3$ substrate on a time scale of a μs. Simultaneously, the temperature of the part of the substrate closest to the illuminated area increases by up to 5 K above 77 K, depending on the laser energy. In the second stage, heat is transferred from the hotter part of the substrate to its cold parts on a time scale of several tens of ms, as measured previously [11]. During all this time temperature gradients are present across the sample. The relatively slow time scale on which the substrate cools is due to its thermal mass, which is much larger than that of the film.

Previously, under homogeneous illumination ($\nabla T \sim 1$ K/cm), we have observed the generation of spontaneous flux during a rapid quench of a superconducting film [3]. The flux appeared faster than the temporal resolution of our SQUID, which is $\sim$10 μs. In the following, we refer to this signal as the "fast" signal. The polarity of the flux from one quench to the next was random. Values of the flux from different quenches followed a Gaussian distribution centered at zero. The width of this distribution increased weakly with the quench rate, a result which is broadly consistent with the Kibble-Zurek scenario.

Under nonhomogeneous illumination, we estimate that ∇T increased to about 300 K/cm for the largest pulse energy used. This is still less than the limit set by the homogeneous criterion [9] of 10^4 K/cm. Under these conditions, the fast signal showed no appreciable change. However, in addition to the fast signal, an unexpected, much larger signal has appeared after a relatively long delay of 1–10 ms (see Fig. 1). This signal was completely absent during measurements using a homogeneous illumination. We point out that the time it takes to cool below T_c is on the order of 1 μs. Consequently, the "slow" signal appears while the film is already in the superconducting state.

The polarity of the nonhomogeneous, slow signal was also random from one quench to the next. Similarly to the fast signal, the amount of flux generated in a given quench followed a Gaussian distribution centered at zero. This is shown in Fig. 2. However, the amount of flux associated with the slow signal is larger than that of the fast signal by an order of magnitude (see also Fig. 3).

After analyzing data acquired using different pulse energies, we found that the amount of spontaneous flux, characterized by the distribution width, increases with the pulse energy. This contrasts the results found under the conditions of uniform illumination (there the distribution width decreased with increasing pulse energy). This is clearly seen in Fig. 3, in which the signal dependence on pulse energy is shown. Note that increasing the pulse energy also increases the thermal gradients generated across the film. Another difference between the fast and slow signals was that at small pulse energy, which was not sufficient to heat the film above T_c, the fast signal disappeared while the slow signal was still there. Finally, measurements were repeated under different external magnetic fields ranging from less than 50 μG up to 60 mG. As Fig. 3 clearly shows, the results do not depend on the external field.

The results at nonhomogeneous conditions point toward two important conclusions. First, as already noted above, increasing the temperature gradients across the film by 2 orders of magnitude (from 1 K/cm up to 300 K/cm) does not change the fast signal. Therefore we conclude that the homogeneous approximation [9] indeed holds, at least for thermal gradients up to $\sim 10^2$ K/cm. Second, the dependence of the slow signal on pulse energy and the long time scale clearly imply that it originates from another mecha-

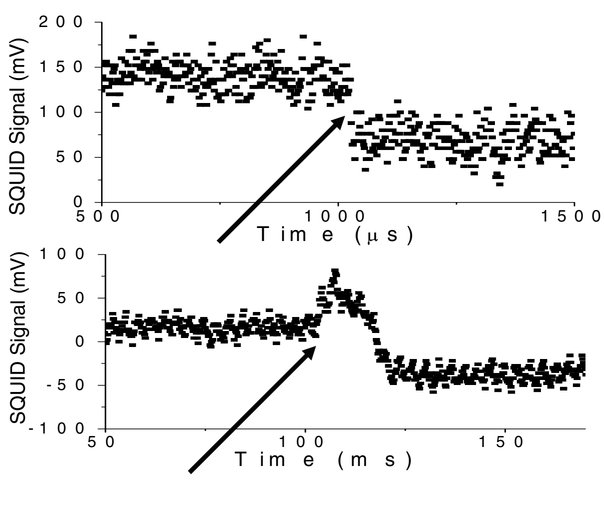

FIG. 1. Typical SQUID signals showing the fast (top trace) and slow (bottom trace) formation of spontaneous flux (note the different scale of the horizontal axes.) Arrows show the time at which the laser pulse was applied.

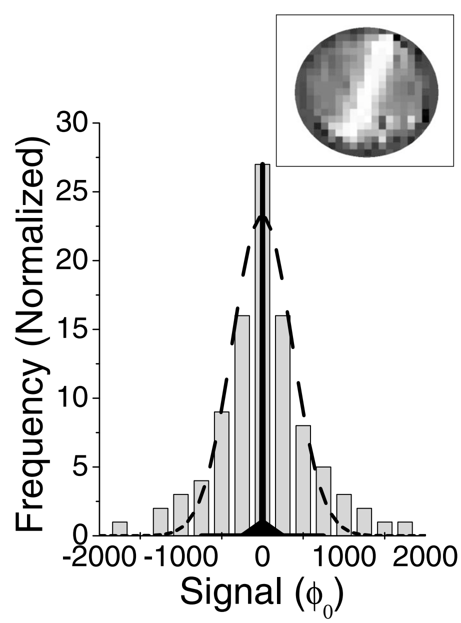

FIG. 2. Typical distribution of spontaneous flux under nonhomogeneous illumination. Solid black bars show the noise distribution, while the dashed curve shows a Gaussian fit to the signal distribution. The inset shows a typical nonhomogeneous illumination profile.

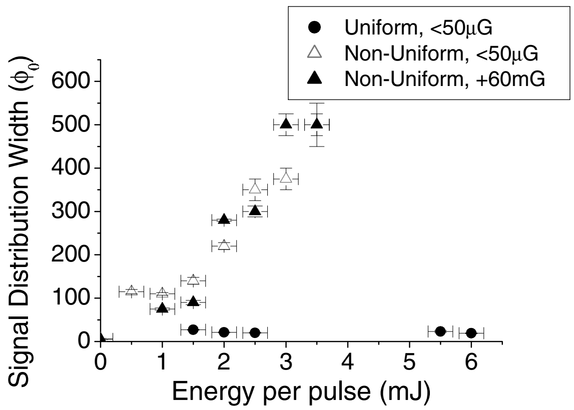

FIG. 3. Signal distribution as a function of pulse energy, showing the difference between homogeneous and nonhomogeneous illumination. Also shown are measurements done at several different external magnetic fields. The error bars are statistical.

nism, rather than the Kibble-Zurek scenario. In the following, we discuss several other mechanisms which may generate magnetic flux, and examine their possible relevance to our observations.

The Hindmarsh-Rajantie model [12] predicts a conversion of thermal energy into magnetic field fluctuations while the sample is in the critical region near T_c. In our experiment, the sample passes through this region in less than 1 μs, while the slow signal develops on a time scale 3 to 4 orders of magnitude slower, 1–10 ms. So, this scenario does not fit with our observations.

Another possibility is a change in the spatial distribution of residual magnetic flux inside the film. Rearrangement of magnetic flux lines can happen during partial illumination of the samples. Magnetic flux can move in or out of the heated part of the film, changing the magnetic flux distribution. Redistribution of magnetic flux can then change the actual amount of flux coupled to the SQUID, even though the net change is zero. We investigated this mechanism in separate measurements done at the university of Konstanz, Germany using a magneto-optic system capable of sub ns resolution [13]. We found that redistribution of flux takes place within several ns, which again is inconsistent with the time scale of our slow signal. In addition, redistribution of flux should depend on the ambient field, which is not borne by our data.

Several theory papers [14,15] proposed that flux can be generated by an instability of a propagating normal-superconducting phase boundary front, which indeed is present in our samples as the film cools after a nonhomogeneous heating pulse. If this mechanism is viable, it should act during less than 1 μs after the heating pulse, since at later times the entire film cools back into the superconducting state and the front disappears. Again, this is 3 orders of magnitude faster than the time after which the slow signal is observed.

Spontaneous flux can be formed at large angle grain boundaries [1,16] as a consequence of the *d*-wave symmetry of the order parameter. Our samples are epitaxial thin films, in which large angle grain boundaries are absent. Hence, this mechanism cannot explain the origin of our signal.

One clue as to the origin of the effect comes from the observation that the temperature gradients across the sample relax on the same time scale as the time over which the slow signal develops. Therefore it is natural to associate it with some thermoelectric effect. This association would also be consistent with the size of the effect increasing with the energy deposited in the film. Thermo-electric effects (the Seebeck effect or the Nernst effect [17]) can generate flux lines as a result of superconducting currents in the film.

In superconductors, the Nernst effect is a result of the motion of flux lines along the thermal gradient. Clearly, this effect depends on the ambient magnetic field. Since we see no such dependence, we conclude that the Nernst effect does not explain our measurements.

Regarding the Seebeck effect in a superconductor, thermal gradients produce a counterflow of normal quasiparticles and Cooper pairs. The net electric current is zero [18]. However, as noted by Ginzburg [19], in some cases such thermoelectric currents can generate magnetic flux. One example is the anisotropic thermoelectric effect [19], in which the supercurrent and the normal current are not colinear and form a current loop. This happens if the Seebeck coefficient is anisotropic and the direction of the thermal gradient is not parallel to one of the superconductor's symmetry axes. Then, the superconducting countercurrent does not exactly cancel the normal current at every point of the film, hence generating a nonzero magnetic flux. Measurements done by Subramaniam *et al.* [20] show that for *untwinned* YBCO crystals, thermoelectric properties are indeed anisotropic. However, our films are twinned, so there is no anisotropy between the **a** and **b** directions (parallel to the surface of the film). Under a temperature gradient of 300 K/cm, we estimate the thermoelectric current $I \sim 5 \times 10^{-5}$ A. This estimate is based on the measured thermal coefficients [20].

If the spontaneous flux were generated via a linear thermoelectric effect, we would expect the polarity of the flux generated to be the same in each measurement, since the temperature gradient in the sample is nominally the same. Since the polarity of the measured flux is random in each measurement, this suggests that perhaps an instability occurs.

One well-known example is the plasma "two stream instability" [21]. In a plasma, the Lorentz force between the two opposing electron beams vanishes only if the currents cancel exactly everywhere. With spatial current fluctuations, the cancellation does not hold, resulting in a net repulsive force between the currents. This in turn leads to further separation of the currents, creating a current loop and a magnetic flux. This situation is illustrated in Fig. 4. We remark that the magnitude of the measured flux is consistent with the picture shown in Fig. 4, namely, with

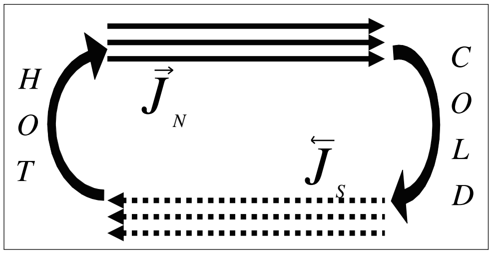

FIG. 4. A schematic picture of the current loop formed by the super and normal thermoelectric currents, separated as a result of the instability.

the current loop having the diameter of the sample. The sense of the current in the loop (clockwise or counter-clockwise) is random, having been determined by the initial fluctuation which separates the current and counter-current. This is in line with our data. In superconductors, the counterflow consists of a normal current opposed by a supercurrent. A high frequency plasma instability in a superconductor was predicted by Kempa *et al.* [22]. Another variant of the ''two stream instability'' was proposed by Bliokh and Shapiro [23], who showed that in the framework of the two-fluid model [24], a uniform superconducting-normal quasiparticle countercurrent is unstable with respect to spatial fluctuations. The analysis reveals that fluctuations in the current density can induce a *low frequency* instability and generate a magnetic field. Denoting the velocities and densities of the normal and superconducting components by $V_{n,s}$, $n_{n,s}$, and taking $n_s V_s = n_n V_n$ and $n_s + n_n = n$, the growth rate of the unstable mode has the form [23]:

$$\omega = \frac{n^2}{n_n n_s} \frac{V_s^2 k^2}{\nu} \tag{1}$$

where ν is the electron relaxation time and k is defined as $k = 2\pi/\Lambda$, with Λ the wavelength of the unstable mode. To obtain a numerical estimate, we determine V_s from $J_s = en_s V_s$, using the thermoelectric current estimated above, $I \sim 5 \times 10^{-5}$ A and the sample cross section. The value of k is determined by taking the size of a fluctuation Λ as λ, the penetration depth, which is the natural scale for flux inhomegeneity. For other quantities in (1), we used the two-fluid model expressions, a charge density n of 10^{21} holes/cm^3, and $\nu \sim 10^{14}$ Hz. Assuming the current flows at the final stage of the experiment near the boundary of the sample, we find $\omega \sim 10^{+3}$ Hz–10^{+4} Hz, which is consistent with the measured experimental growth rate (ranging between 10 Hz–10^3 Hz). Hence, this scenario gives a possible explanation for the origin of the measured spontaneous flux.

In conclusion, we have discovered a new mechanism of spontaneous flux generation in a superconductor quenched through T_c in the presence of a temperature gradient. One mechanism which may be responsible for this new effect is an instability of the thermoelectric current distribution.

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*Corresponding author.
Electronic address: mariel@tx.technion.ac.il

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