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

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Abstract

Magnetic flux can be generated spontaneously in a superconductor during cooldown through the critical region near T_c. We discovered that in the presence of a temperature gradient, an additional mechanism becomes operative, generating spontaneous flux of random sign and a magnitude which increases with the size of the temperature gradient. One interesting suggestion is that flux is generated through some instability of the thermoelectric superconducting-normal quasiparticle counterflow.

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The subject of a order-disorder phase transition occurring under non equilibrium conditions has attracted considerable attention recently. Experiments done in this area are significant with respect to the applicability of the Kibble-Zurek cosmological scenario[1, 2] to condensed matter systems, as well as to the general subject of non-equilibrium statistical mechanics. Basically, the Kibble-Zurek scenario claims that if a system having a complex order parameter is quenched rapidly through the transition temperature into the ordered state, topological defects of the order parameter will be created. In superconductors and superfluid Helium, the topological defects are quantized vortex lines[2].The density of defects is predicted to increase with the quench rate. The Kibble-Zurek scenario[1, 2] was tested in several experimental systems, ranging from superfluid helium[3, 4] and liquid crystals[5, 6] to superconductors[7, 8, 9, 10, 11].

One of the basic assumptions of the Kibble-Zurek scenario is that the phase transition is homogeneous, in the sense 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[12], 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[2].The motivation for our experiment was 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. 11, with the exception of a non-uniform 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[13]. Single pulses (FWHM $\sim$ 10 ns) were used to heat the film. The laser pulse passes through the substrate and illuminates *non-uniformly* 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. 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.

Non-uniform illumination was generated either by using a non-uniform light beam, or by covering some part of the sample. An example of one such arrangement is shown in the inset of figure 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 4mm 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 non-uniform illumination, the film cools down in a two stage process. 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. As a result, 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. 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[11]. The flux appeared faster than the temporal resolution of our SQUID, which is $\sim 10\mu$s. 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, following approximately 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 non-homogeneous 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[12] 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 figure 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 non-homogeneous, "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 Figure 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 figure 3).

After analyzing data aquired 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 figure 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.

Finally, measurements were repeated under different external magnetic fields ranging from less than 50 μG up to 60 mG. As figure 3 clearly shows, the results do not depend on the external field.

The results at non-homogeneous conditions point towards 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[12] 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 mechanism, 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[14] 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. Re-arrangement 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. Re-distribution 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 [15]. We found that re-distribution of flux takes place within several ns, which again is inconsistent with the time scale of our slow signal. In addition, re-distribution of flux should depend on the ambient field, which is not borne by our data.

Magnetic 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 non-homogeneous heating pulse. This idea was proposed in several theoretical papers[16, 17]. This mechanism should be operative 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 order of magnitude faster than the time after which the slow signal is observed.

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[18]) 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 counter-flow of normal quasiparticles and Cooper pairs. The net electric current is zero[19]. However, as noted by Ginzburg[20], in some cases such thermo-electric currents can generate magnetic flux. One example is the anisotropic thermo-electric effect[20], in which the supercurrent and the normal current are not co-linear 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 non-zero magnetic flux. Measurements done by Subramaniam et al.[21] 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 thermo-electric current $I \sim 5 \times 10^{-5}$A. This estimate is based on the measured thermal coefficients[21].

If the spontaneous flux was 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"[22]. 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 current loops (see the inset of figure 4). Such instability can generate a magnetic flux. In superconductors, the situation is slightly different, as the counterflow consists of a normal current opposed by a supercurrent, and is described by the equations of superconducting dynamics. In the framework of the two-fluid model[23] one can show that a uniform superconducting-normal quasiparticle countercurrent is unstable with respect to spatial fluctuations[24]. 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:

$$\omega = \frac{(n_n + n_s)^2}{n_n n_s} \frac{V_s^2 k^2}{\nu}; \tag{1}$$

where ν is the electronic relaxation time and k is the wavevector of the unstable mode. To obtain a numerical estimate, we take the size of a fluctuation as ξ, the coherence length. This choice fixes $k = 2\pi/\xi$. V_s is determined from $J_s = e n_s V_s$, using the thermoelectric current estimated above, $I \sim 5 \times 10^{-5}$A and the sample cross section. 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. A plot of ω as a function of temperature is shown in Fig 4. The experimental values agree with Eq. (1) only very close to T_c. This is inconsistent with the long time which passes until the appearance of the signal. However, we feel that the basic idea of a thermoelectric instability is intriguing and deserves further study.

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. Among the various mechanisms which may be responsible for this effect, an instability of the thermoelectric current seems an intriguing possibility.

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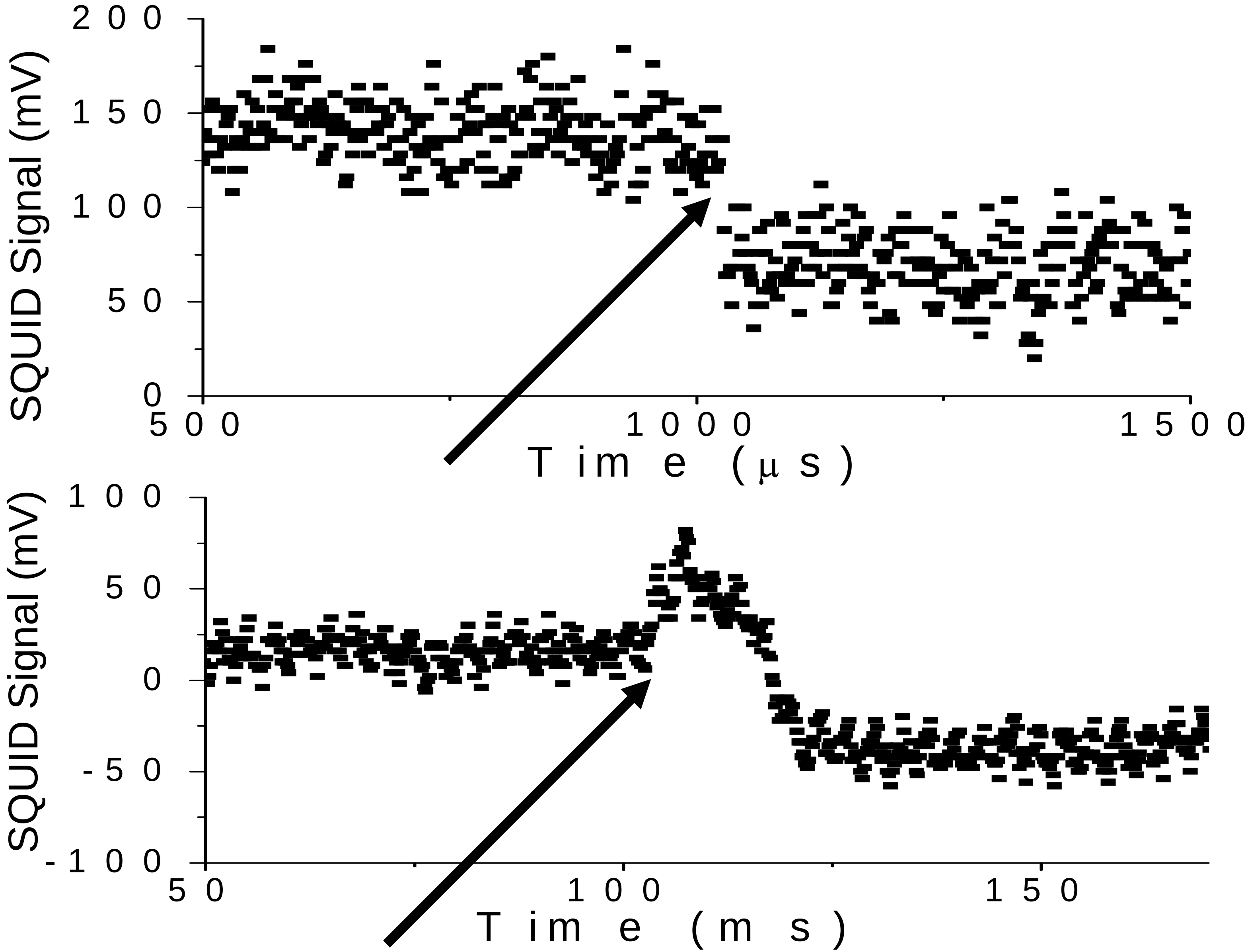
FIG. 1: Typical SQUID signals showing the fast (top trace) and slow(bottom trace) formation of spontaneous flux (note the different scale of the horizontal axes.) Arrows show the time at which the laser pulse was applied.

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

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

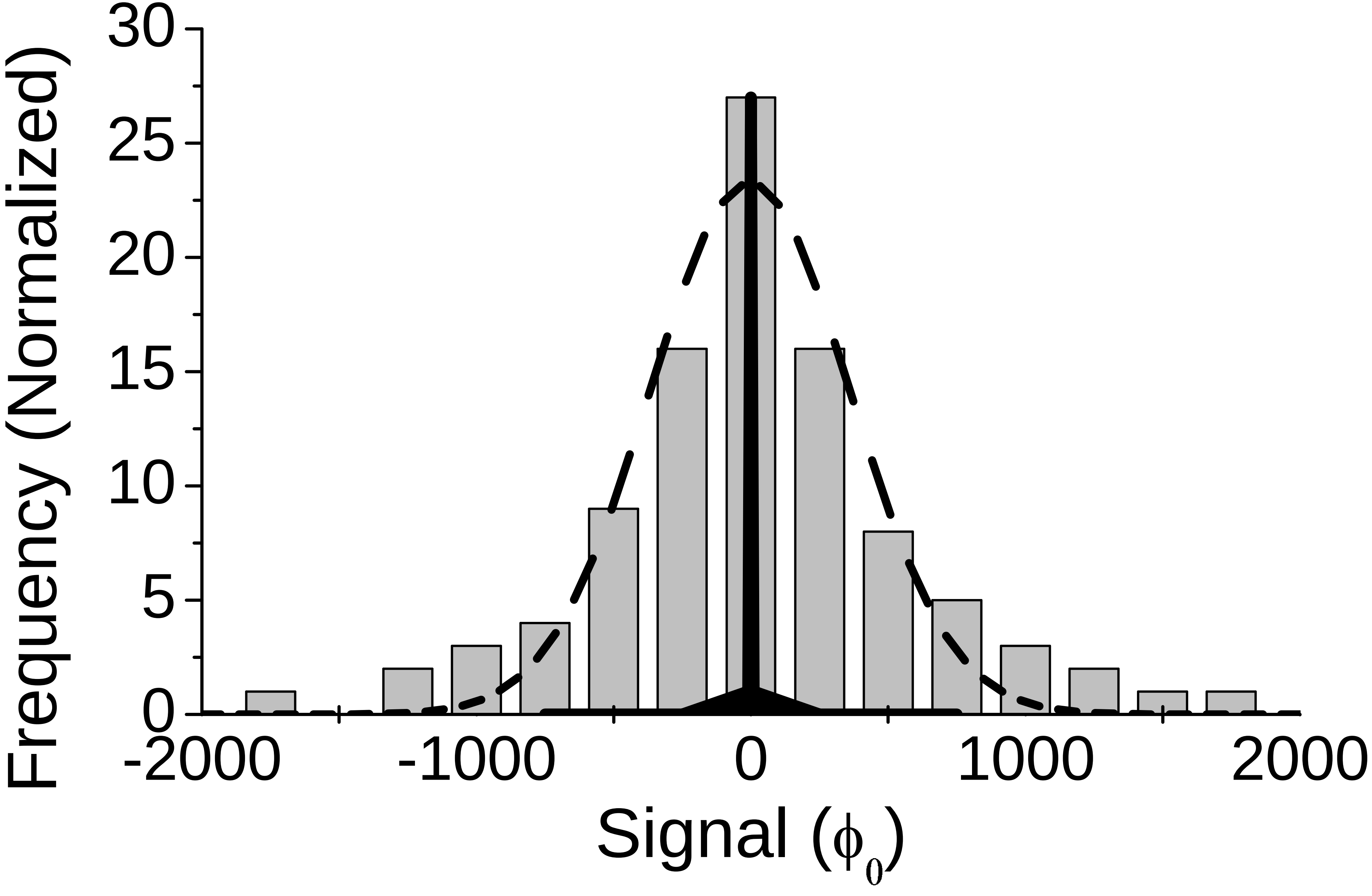
FIG. 4: Instability growth rate as a function of temperature. The solid line is the theoretical prediction (Eq. 1). The dashed lines represent the lower and upper limits on the growth rate, extracted from our measurements. The inset shows a schematic picture of the current loop formed by the super and normal thermoelectric currents, separated as a result of the instability.

Figure No. 1



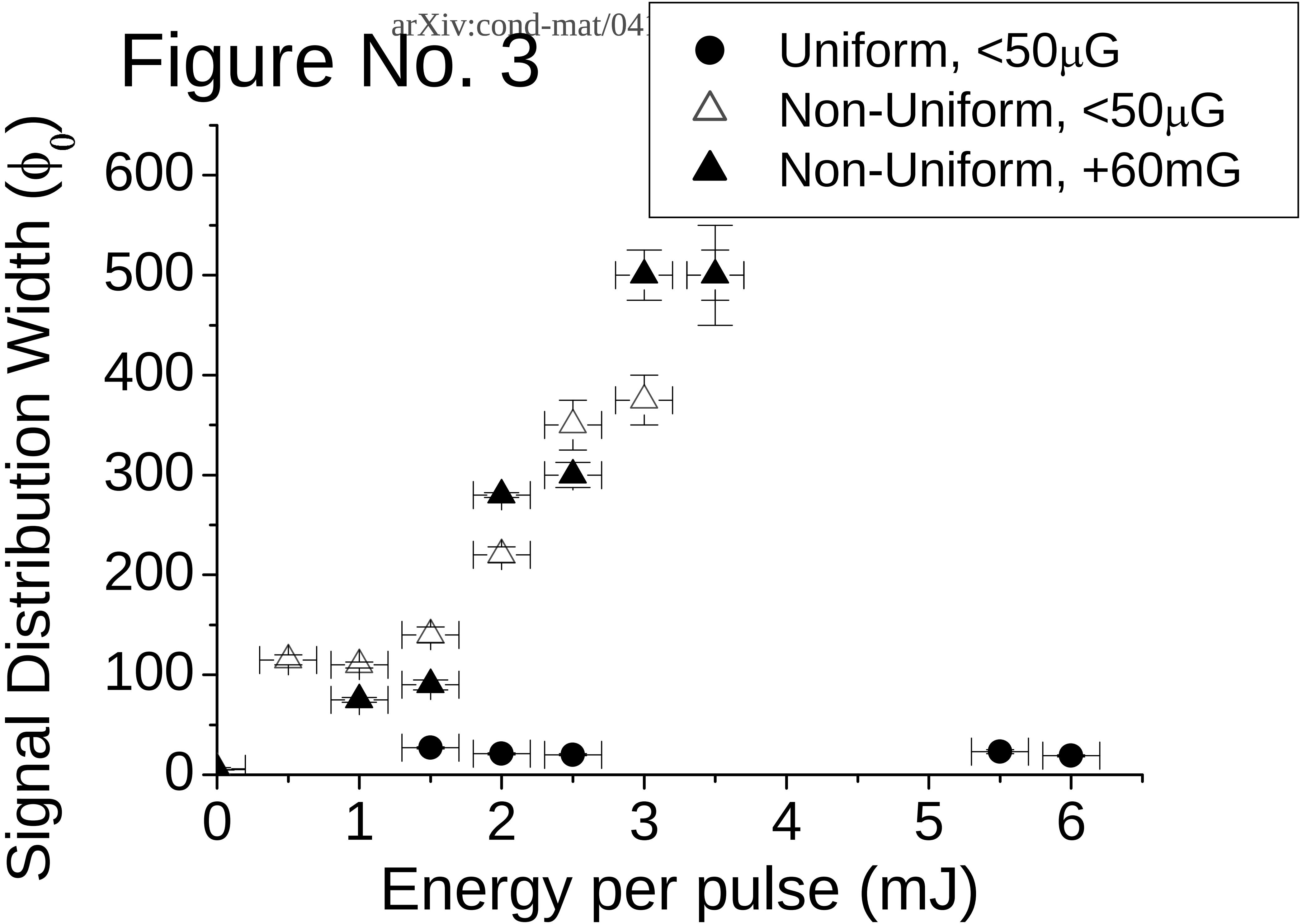
SQUID Signal (mV)
200
150
100
50
0
500
1000
1500
Time (μs)
SQUID Signal (mV)
100
50
0
-50
-100
50
100
150
Time (ms)

Figure No. 2



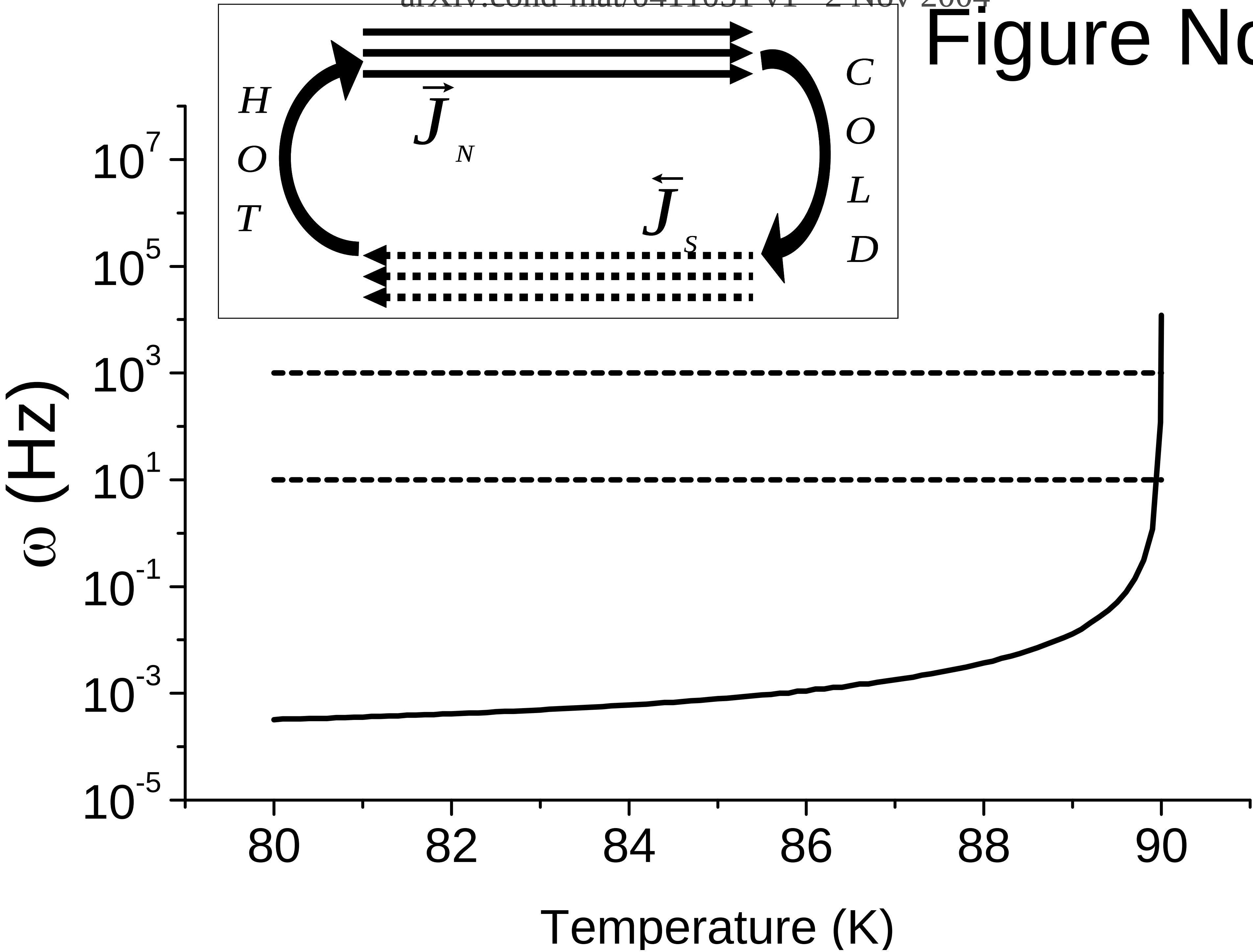
Frequency (Normalized)
30
25
20
15
10
5
0
-2000
-1000
0
1000
2000
Signal (ϕ_0)

Figure No. 3



Uniform, <50μG
Non-Uniform, <50μG
Non-Uniform, +60mG
Signal Distribution Width (ϕ_0)
0
100
200
300
400
500
600
0
1
2
3
4
5
6
Energy per pulse (mJ)

Figure No. 4


H
O
T
C
O
L
D
$\vec{J}_N$
$\overleftarrow{J}_S$
10^7
10^5
10^3
10^1
10^{-1}
10^{-3}
10^{-5}
ω (Hz)
80
82
84
86
88
90
Temperature (K)