Zurek-Kibble Mechanism for the Spontaneous Vortex Formation in Nb-Al/Al_{ox}/Nb Josephson Tunnel Junctions: New Theory and Experiment

R. Monaco,[1,*] J. Mygind,[2,†] M. Aaroe,[2] R. J. Rivers,[3,‡] and V. P. Koshelets[4,§]

[1]Istituto di Cibernetica del C.N.R., 80078, Pozzuoli, Italy and Unitá INFM-Dipartimento di Fisica, Universitá di Salerno, 84081 Baronissi, Italy

[2]Department of Physics, B309, Technical University of Denmark, DK-2800 Lyngby, Denmark

[3]Blackett Laboratory, Imperial College London, London SW7 2AZ, United Kingdom

[4]Institute of Radio Engineering and Electronics, Russian Academy of Science, Mokhovaya 11, Building 7, 125009, Moscow, Russia

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New scaling behavior has been both predicted and observed in the spontaneous production of fluxons in quenched Nb-Al/Al_{ox}/Nb annular Josephson tunnel junctions (JTJs) as a function of the quench time, τ_Q. The probability f_1 to trap a single defect during the normal-metal–superconductor phase transition clearly follows an allometric dependence on τ_Q with a scaling exponent $\sigma = 0.5$, as predicted from the Zurek-Kibble mechanism for *realistic* JTJs formed by strongly coupled superconductors. This definitive experiment replaces one reported by us earlier, in which an idealized model was used that predicted $\sigma = 0.25$, commensurate with the then much poorer data. Our experiment remains the only condensed matter experiment to date to have measured a scaling exponent with any reliability.

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Because phase transitions take place in a finite time, causality guarantees that correlation lengths remain finite, even when the transitions are continuous. The Zurek-Kibble (ZK) scenario [1–3] proposes that such transitions take effect as fast as possible, i.e., the domain structure after the quenching of the system initially reflects the causal horizons. As a result, once the transition is implemented, correlation lengths scale as (positive) powers of the quench time τ_Q (inverse quench rate). This proposal is amenable to direct testing for transitions whose domain boundaries carry topological charge. Such is the case for Josephson tunnel junctions (JTJs), where the topological charge is a fluxon, i.e., a supercurrent vortex carrying magnetic flux $\Phi_0 = h/(2e)$ in the plane of the oxide layer between the two superconductors that make up the JTJ. In the case considered in this Letter of annular JTJs, obtained by the superposition of two superconducting rings, it corresponds to a magnetic flux that threads just one of the two rings an odd number of times.

In 2000 an idealized model was proposed [4] to test the ZK scenario for JTJs, assuming that causal horizons are constrained by the velocity of electromagnetic waves in the JTJ, the Swihart velocity [5,6]. As a result, the probability f_1 for spontaneously producing one fluxon in the thermal quench of a symmetric annular JTJ of circumference C was predicted to scale as

$$f_1 \simeq \frac{C}{\bar{\xi}} = \frac{C}{\xi_0}\left(\frac{\tau_Q}{\tau_0}\right)^{-\sigma}. \quad (1)$$

In Eq. (1), $\bar{\xi}$ is the Zurek-Kibble causal length, the correlation length of the relative phase angle at the time of defect formation. It is defined through Eq. (1) in terms of the cold correlation length ξ_0, the relaxation time τ_0 of the long wavelength modes, and the quench time τ_Q, in turn defined by: $T_C/\tau_Q = -(dT/dt)_{T=T_C}$. Equation (1) holds for $C < \bar{\xi}$. Under the assumptions of (a) weak coupling of the superconductors and (b) exact critical slowing of the Swihart velocity at the critical temperature $T = T_c$, we predicted $\sigma = 0.25$ [4].

In 2001 our first proof-of-principle experiment [7] was performed, to test Eq. (1). The experiment consisted of taking an annular JTJ isolated from its surroundings and making it undergo a forced phase transition by heating it above its superconducting critical temperature and letting it cool passively back towards the liquid-He temperature. Once the thermal cycle is over, the junction *I-V* curve is inspected and any trapped fluxon can be detected by the appearance of current peaks at discrete voltages in the *I-V* characteristic of a JTJ. By detecting the voltage position of these peaks the number of trapped fluxons (or antifluxons) can be determined.

The experiment of Ref. [7] was remarkably successful, as the only experiment of the several [8–17] previously performed on condensed matter systems sensitive enough to show quantitative ZK scaling behavior, although qualitative support for scaling had been demonstrated with ^{3}He and high-T_c superconductors. However, this preliminary experiment suffered from severe restrictions, in particular, a limited range of cooling rates, statistically poor data, and above all insufficient shielding of the earth's magnetic field with the possibility of systematic errors. The outcome was σ commensurate with 0.25.

In this Letter we shall present results from a new experiment designed to circumvent these problems, that we shall discuss later. Our experiment shows extremely reliable scaling behavior of the form (1), but with $\sigma = 0.5$ to high accuracy. This is obviously at total variance with our earlier prediction. However, as we shall show, the value

$\sigma = 0.25$ depended critically on both assumptions (a) and (b) cited earlier being *exactly* satisfied. For realistic JTJs these assumptions are only *approximately* satisfied in the immediate vicinity of $T = T_c$. Since defects form very close to T_c, we shall see that this is sufficient for the value of σ to jump from 0.25 to $\sigma = 0.5$. Thus, rather than its negation, our experiment arguably provides an even more robust demonstration of the ZK scenario.

To reiterate, the theory in Ref. [4] was developed for JTJs whose electrodes are *weak coupling* superconductors; for such JTJs the temperature dependence of the critical current density $J_c(T)$ is given by the Ambegaokar-Baratoff equation [18]:

$$J_c(T) = \frac{\pi}{2}\frac{\Delta(T)}{e\rho_N}\tanh\frac{\Delta(T)}{2k_BT}, \tag{2}$$

where $\Delta(T)$ is the superconducting gap energy and ρ_N is JTJ normal resistance per unit area squared. Equation (2) provides a linear decrease of J_c near T_c:

$$J_c[T(t)] = \alpha J_c(0)\left(1 - \frac{T}{T_c}\right) \simeq \alpha J_c(0)\frac{t}{\tau_Q}, \tag{3}$$

where $T(t = 0) = T_c$ and the dimensionless quantity α is approximately equal to $2\Delta(0)/k_BT_C = 3.5$.

However, our JTJs are based on Nb, a *strong-coupling* superconductor, for which Eq. (2) is not necessarily valid. In practice, high quality and reproducible barriers are achieved by depositing a thin Al overlay onto the Nb base electrode which will be only partially oxidized, leaving a Nb-Al bilayer underneath having a non-BCS temperature dependence of the energy gap and of the density of states. The proximity effect in Nb-Al/Al_{ox}/Nb JTJs has been extensively studied and it is known to influence the electrical properties of the junctions, such as the current-voltage characteristic and the temperature dependence of the critical current density. Specifically, the proximity effect in superconductor–normal-metal–insulator–superconductor junctions can lead to dominance by an otherwise subdominant temperature dependence of the critical current density [19] in the vicinity of T_c of the form

$$J_c(T(t)) \simeq \alpha' J_c(0)\left(1 - \frac{T}{T_c}\right)^2 = \alpha' J_c(0)\left(\frac{t}{\tau_Q}\right)^2, \tag{4}$$

where α' is a constant depending intricately on the degree of proximity. The last equation models the tail shaped dependence of J_c vs T near T_c; it has been theoretically derived and experimentally confirmed by Golubov *et al.* [20] in 1995. Rephrasing the arguments of Ref. [4] with Eq. (4) replacing Eq. (3), the Josephson penetration depth $\lambda_J(T)$, which plays the role of system equilibrium coherence length $\xi(T)$, near T_c diverges linearly with time

$$\xi[T(t)] = \lambda_J(T(t)) = \frac{\xi_0\tau_Q}{t}, \tag{5}$$

where

$$\xi_0 = \sqrt{\frac{\hbar}{2e\mu_0 d_s\alpha' J_c(0)}}, \tag{6}$$

d_s being electrode thickness.

$\dot{\xi}(t) = d\xi(t)/dt < 0$ measures the rate at which the defects contract, i.e., the speed of interfaces between ordered and disordered ground states. Since $\dot{\xi}(t)$ decreases with time $t > 0$, the *earliest* possible time t at which defects could possibly appear is determined by causality,

$$\dot{\xi}(\bar{t}) = -\bar{c}(\bar{t}), \tag{7}$$

where $\bar{t}$ is the causal time and $\bar{c}$ is the Swihart velocity.

As we said, in Ref. [4] we had also assumed that the Swihart velocity vanished at T_c. For realistic JTJs, this is not so; it just becomes very small. Swihart [5] has demonstrated that for a thin-film superconducting strip transmission line the solution for the velocity varies continuously as one passes through the critical temperature into the normal state. As a result, we assume $\bar{c}(t) = \bar{c}_{nn}$ near the transition temperature where $\bar{c}_{nn}$ is the speed of light in a microstrip line made of normal metals. In the case of a microstrip line made by two electrodes having the same thickness d_s and the same skin depth δ, with $d_s \ll \delta$, separated by a dielectric layer of thickness d_{ox} and dielectric constant ϵ, $\bar{c}_{nn} \simeq (2/\delta)\sqrt{d_{ox}d_s/\epsilon\mu}$ [5]. Its value depends on the temperature very weakly, but depends on the frequency f through $\delta = \sqrt{\rho/\pi\mu f}$, ρ being the normal-metal residual resistivity.

The match $\bar{c}(T_c) = \bar{c}_{nn}$ is certainly realistic and we still have approximate critical slowing down insofar as $\bar{c}_{nn}$ is much smaller than the zero temperature Swihart velocity $\bar{c}_0 \simeq \sqrt{d_{ox}/2\lambda_{L0}\epsilon\mu}$, i.e., when the zero temperature London penetration depth $\lambda_{L0} \ll \delta^2/d_s$. For 300 nm thick Nb electrodes ($\rho = 3.8\ \mu\Omega\,\mathrm{cm}$ and $\lambda_{L0} = 90$ nm), $\delta \simeq$ 1 mm at say $f = 10$ kHz, so the last inequality is fully satisfied. At the same frequency, for a value of the specific barrier capacitance $c_s = \epsilon/d_{ox} = 0.027\ \mathrm{F/m^2}$ typical of low current density Nb-Al/Al_{ox}/Nb JTJs, we get $\bar{c}_{nn} = 6 \times 10^3$ m/s and $\bar{c}_0 = 1.6 \times 10^7$ m/s [21].

The solution of the causality equation Eq. (7) with a nonvanishing Swihart velocity yields: $\bar{t} = \sqrt{\xi_0\tau_Q/\bar{c}_{nn}}$, again discretely different from its idealized counterpart [4]. Inserting the value of $\bar{t}$ into Eq. (5) we obtain the new Zurek length $\bar{\xi}$

$$\bar{\xi} = \xi(\bar{t}) = \sqrt{\xi_0\tau_Q\bar{c}_{nn}} = \xi_0\left(\frac{\tau_Q}{\tau_0}\right)^{1/2}, \tag{8}$$

where $\tau_0 = \xi_0/\bar{c}_{nn}$ ($\tau_0 = O(1$ ns). We reach the important conclusion for realistic JTJs that the probability f_1 for spontaneously producing one fluxon in the quench is still predicted to scale with the quench time τ_Q according to Eq. (1), but the critical exponent is now $\sigma = 0.5$, rather than $\sigma = 0.25$. Detailed calculations will be reported elsewhere [22].

However, it is worth observing that by varying τ_Q in the experimentally achieved four-decade range 1 ms $< \tau_Q <$ 10 s, we get 10 μs $< \bar{t} <$ 1 ms that is always much larger than τ_0; it means that by the time the Josephson phase *freezes* the Josephson effect is well established. Further, in the same τ_Q range the normalized freezing temperature $\bar{T}/T_C$ at which the defect is formed is $0.99 < \bar{T}/T_C <$ 0.9999. It would be really hard, if not impossible, to measure the temperature dependence of J_c and $\bar{c}$ so close to T_C. We have then to resort to theoretical predictions.

The new experiment has a faster and more reliable single heating system, obtained by integrating a meander line 50 μm wide, 200 nm thick, and 8.3 mm long Mo resistive film in either ends of the 4.2 mm $\times$ 3 mm $\times$ 0.35 mm Si chip containing the Nb/AlOx/Nb trilayer JTJs. These resistive elements have a nominal resistance of 50 Ω at LHe temperatures and, due to their good adhesion with the substrate, are very effective in dissipating heat in the chip. In fact, voltage pulses a few μs long and a few volts high applied across the integrated heater provided quench times as low as 1 ms, that is more than 2 orders of magnitude smaller than for the previous situation [7]. Further, automatization of thermal cycles was implemented that allowed for much more robust statistics to be achieved. At the end of each thermal quench the junction *I-V* curve is automatically stored and an algorithm has been developed for the detection of the trapped fluxons. Finally, all the measurements have been carried out in a magnetic and electromagnetically shielded environment. The low frequency magnetic shielding was achieved by using μ-metal, cryoperm, and lead cans. During the quench the JTJ was also electrically isolated.

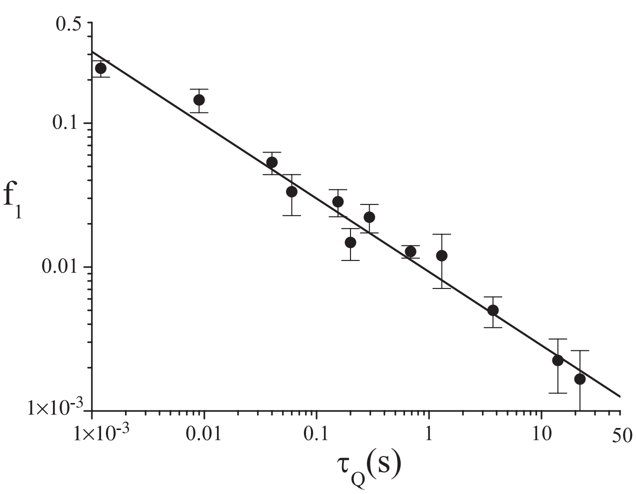

FIG. 1. Log-log plot of the measured frequency f_1 of trapping single fluxons versus the quenching time τ_Q. Each point corresponds to many thermal cycles. The vertical error bars gives the statistical error while the relative error bars in τ_Q amounting to $\pm 10\%$ are as large as the dots' width. The solid line is the fit to an allometric relationship $f_1 = a\tau_Q^{-b}$ which yields $a =$ 0.0092 $\pm$ 10% (taking τ_Q in seconds) and $b = 0.51 \pm 6\%$.

The experimental results reported here are restricted to one of two annular JTJs with similar geometry to the sample used in the earlier experiment ($C = 500$ μm and $d_s = 300$ nm), but with about 50 times lower critical current density ($J_c(0) \simeq 60$ A/cm^2): this leads to a value of $\xi_0 = 17$ μm (we have assumed $\alpha' = \alpha = 3.5$ in Eq. (6)). An extensive description of the chip layout, the experimental setup and the quenching data will be given elsewhere [22] while the details of the fabrication process can be found in Ref. [23]. The quench time τ_Q was continuously varied over more that 4 orders of magnitude (from 20 s down to 1 ms) by varying the helium exchange gas pressure inside the vacuum can and/or the width and the amplitude of the voltage pulse across the integrated resistive element.

Figure 1 shows on a log-log plot the measured frequency $f_1 = n_1/N$ of single fluxon trapping, obtained by quenching the sample N times for each value of a given quenching time τ_Q, n_1 being the number of times that the inspection of the low temperature JTJ current-voltage characteristics at the very end of each thermal cycle showed that one defect was spontaneously produced. N ranged between 100 and 2600 and n_1 was never smaller then 10, except for the rightmost point for which $n_1 = 3$ (and $N = 1800$). The sample has undergone a total of more than 100 000 thermal cycles without any measurable change of its electrical parameters. The vertical error bars gives the statistical error $f_1/\sqrt{n_1}$. The measurement of τ_Q by fitting solutions to the heat equation follows that of [7] in its high accuracy. The relative error bars in τ_Q amounting to $\pm 10\%$ are as large as the dot's width. To test Eq. (1), we have fitted the data with an allometric function $f_1 = a\tau_Q^{-b}$, with a and b as free fitting parameters. A linear regression of $\log f_1$ vs $\log\tau_Q$, represented by the continuous line in Fig. 1, yields $b = 0.51 \pm 6\%$, in excellent agreement with the predicted value 0.5. The same fit gives $a = 0.0092 \pm$ 10% (taking τ_Q in seconds) that is 6–7 times larger than the predicted value $C/\sqrt{\xi_0 \bar{c}_{nn}}$ (with $C = 500$ μm, $\xi_0 =$ 17 μm and $\bar{c}_{nn} = 6 \times 10^3$ m/s). As a bound we only expect agreement in the overall normalization a to somewhat better than an order of magnitude. Empirically, the different condensed matter experiments have shown that the ratio $a_{\mathrm{observed}}/a_{\mathrm{predicted}}$ varies widely from system to system; $O(1)$ for superfluid ^{3}He [8,9], very small for high-T_c superconductors [13]. We point out that in our case the value of the prefactor a is dependent, although weakly, on the choice of α' and f, being $C/\sqrt{\xi_0 \bar{c}_{nn}} \propto (\alpha'/f)^{1/4}$. The choice $f = 10$ kHz was determined assuming that $1/a^2 \simeq 12$ kHz is the characteristic frequency of our system at the time of the thermal quench. As we noted earlier, the new scaling exponent $\sigma = 0.5$, obtained by applying causality arguments to *realistic* JTJs, is twice as large than that observed earlier [7], also in samples made with the same Nb-Al/Al$_{\mathrm{ox}}$/Nb technology. The reason for this discrepancy resides in the fact that at that time we were unaware of the high sensitivity of f_1 to external magnetic fields which, although small, were most likely present. In the present experiment much care has been devoted both to avoid magnetic or current carrying materials in the cryoprobe during the quench and to screen the chip environment from 50 Hz noise as well as from dc magnetic field.

The data of Fig. 1 resolve another issue. It has been disputed that the Swihart velocity, which gives the behavior above, is the relevant velocity for field ordering along the JTJ oxide. If we had taken the relevant velocity to be that of phase ordering in the individual superconductors, as invoked by Zurek [1,2] when considering the spontaneous flux produced on quenching annuli of simple superconductors, at the same level of approximation we would have predicted $b = 0.25$ and the prefactor orders of magnitude larger.

We have not observed the change in behavior predicting f_1 to change from the linear behavior with $C/\bar{\xi}$ of Eq. (9) to a random walk behavior in the phase,

$$f_1 \simeq \sqrt{C/\bar{\xi}} = \sqrt{\frac{C}{\xi_0}\left(\frac{\tau_Q}{\tau_0}\right)^{-\sigma/2}}, \qquad (9)$$

once f_1 is sufficiently large. A first guess would suggest that the transition from Eq. (1) to Eq. (9) would occur when $f_1 \approx 1$. However, future experiments to be carried out on ring-shaped JTJs having the circumference larger than that of the sample reported in this paper and with τ_Q in the same range of this experiment should clearly reveal the transition from Eq. (1) to Eq. (9).

In summary, we see this experiment as providing strong corroboration of Zurek-Kibble scaling over a wide range of quenching time τ_Q in accord with our predictions for Nb-Al/Al_{ox}/Nb JTJs. As such, it replaces the experiment reported in Ref. [7] by being more realistic theoretically and more sophisticated experimentally. We stress that this experiment is the *only* one to date to have confirmed the Zurek-Kibble scaling exponent for a condensed matter system. Further experiments can be devised to investigate the transition to the random walk regime and the effect of the thermal gradients. Such experiments are in the process of being performed.

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*Electronic address: roberto@sa.infn.it
†Electronic address: myg@fysik.dtu.dk
‡Electronic address: r.rivers@imperial.ac.uk
§Electronic address: valery@hitech.cplire.ru

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