

Granular Josephson and quantum interference effects in HTC ceramic superconductors

by C. E. Gough

Josephson effects in high- T_c superconductors are briefly reviewed, with specific reference to granular ceramic materials and SQUID device applications. It is suggested that the inductance associated with intergranular current loops may play an important role, even in determining the bulk superconductivity properties, as in weak-link superconducting rings. Evidence for quantum interference effects within intergranular current loops is presented. In ultra-low fields, the observed temperature dependence of thermally activated flux creep cannot be described by a simple granular superconductor model of equally spaced pinning centers, but would seem to imply a hierarchy of pinning sites of variable strength. The development of liquid-nitrogen-cooled rf and dc SQUIDs is described, and the noise levels currently achieved are presented.

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1. Introduction

The discovery of high-temperature superconductivity in cuprate perovskite compounds [1] has enabled macroscopic quantum effects to be demonstrated at liquid-nitrogen temperatures for the first time. The first demonstration of quantum physics in HTC superconductors was the Saclay measurement of the inverse ac Josephson effect [2], followed rapidly by the Birmingham flux quantum determination [3].

These measurements demonstrated the importance of Josephson coupling of superconducting electrons [4] between the grains of granular HTC superconductors. Such coupling dominates the low-field properties of HTC superconductors. In particular, the strength of the Josephson coupling between grains determines the maximum supercurrent that can be supported by the bulk material. Weak links, possibly associated with internal domain structures or twin boundaries, also appear to play an important role in determining the superconducting properties of individual grains and single crystals [5].

As weak links are intrinsic features of any ceramic superconductor, understanding their properties and their dependence on microstructure is central to the problem of optimization of critical currents. In this brief review, we consider the influence of Josephson currents on the microscopic electrical and magnetic properties of HTC

ceramic superconductors and consider, in particular, the influence of flux quantization on bulk properties and the development of SQUIDS using bulk ceramic superconductors.

2. The Josephson effect

Consider the quantum-mechanical coupling of superconducting electrons between two superconducting grains separated by a thin nonsuperconducting barrier. In practice, such a barrier might be a naturally occurring grain boundary, a nonsuperconducting region of amorphous, nonstoichiometric or second-phase material between two ceramic grains, or a twin boundary or crystallographic domain wall within a grain. As the superconducting coherence length in HTC materials is so very short (for YBCO, $\lesssim 5 \text{ \AA}$ in the c -direction and $\lesssim 30 \text{ \AA}$ in the a - b plane [6]), lattice defects or changes in stoichiometry, even on an atomic scale, can give rise to significant local depression of the superconducting order parameter [5].

The macroscopic superconducting state on either side of a junction can be described by a wavefunction varying as $\exp(iS)$, where S is a phase factor. When superconducting electrons cross a barrier, $I = I_c \sin \theta$ and $d\theta/dt = 4\pi eV/h$, where θ is the phase difference and V the voltage across the junction [4, 7]. These are the well-known Josephson current and voltage phase relations.

When $I < I_c$, pairs of superconducting electrons can pass across the weak link with no voltage appearing across it (the dc Josephson effect). For an ideal tunnel junction, $I_c = (\pi/2)\Delta/eR$ [8], where R is the resistance across the junction in the normal state and Δ is the superconducting energy gap. This expression allows an estimate to be made of the maximum critical current density in a ceramic superconductor, $J_c = I_c/a^2 \sim \Delta/epa$, where ρ is the bulk resistivity and a the average intergrain spacing. Assuming a value for $\Delta/e \sim 20 \text{ mV}$, a resistivity of $100 \mu\Omega\text{-cm}$, and a grain size of $10 \mu\text{m}$, a maximum $J_c \sim 2 \times 10^5 \text{ A-cm}^{-2}$ is obtained, which is an order of magnitude larger than has yet been achieved in sintered material.

The IBM group at Yorktown Heights has recently published a series of beautiful papers [9] reporting measurements of Josephson junction tunneling between and within single-crystal grains in epitaxial thin films of YBCO on oriented substrates. The critical current across grain boundaries was usually significantly less than that measured in the grains on either side of a boundary, confirming that it is largely the weak intergrain coupling that limits the supercurrent in ceramic materials.

As soon as I_c is exceeded, a voltage is developed, resulting in a sinusoidal variation of current through the junction, $I = I_c \sin [2\pi(2eV/h)t]$, providing a source of microwave radiation with frequency $f = 2eV/h = 484 \text{ MHz}/\mu\text{V}$. This corresponds to the radiation of photons of energy hf equal to the gain in energy $2eV$ as pairs of electrons cross the junction.

When a Josephson junction is exposed to microwave radiation of frequency f , steps appear in the I/V characteristics at voltages which are multiples of $hf/2e$. This inverse ac Josephson effect provided the first experimental confirmation of the pairing of electrons in high- T_c ceramic superconductors [2]. Similar microwave steps have subsequently been observed in ceramic samples with weak links produced by crack junctions [10] and in material thinned down to form a narrow constriction region [11]. The ac Josephson effects are assumed to occur across the weak links between the grains of the granular ceramic material.

3. Superconducting weak-link rings

Within a superconducting ring containing a weak link across which Josephson tunneling can occur, the requirement that the phase S must everywhere be single-valued results in a direct relationship between θ , the phase difference across the weak link, and the magnetic flux contained within the ring, $\theta = 2\pi\varphi/\varphi_0$, where $\varphi_0 = h/2e = 2.07 \times 10^{-15} \text{ T-m}^2$ is the flux quantum. The $2e$ in the denominator assumes the pairing of electrons in the superconducting state. The current flowing around the ring can then be expressed as $I = I_c \sin(2\pi\varphi/\varphi_0)$. When an external flux, φ_{ext} , is applied—for example, by placing a current-carrying coil inside the ring—a shielding supercurrent is induced such that $\varphi_{\text{ext}} = \varphi_0 + LI_c \sin 2\pi\varphi/\varphi_0$, where L is the inductance of the ring.

The magnetic behavior of the ring then depends on the value of $\beta = 2\pi LI_c/\varphi_0$. When $\beta < 1$, the magnetic properties of the ring are reversible. However, when $\beta > 1$, the magnetic properties become irreversible, and energy is dissipated on cycling the external field around a magnetic hysteretic loop. This energy loss is a periodic function of any applied static field with the periodicity of the flux quantum. This is the basis of operation of rf and dc SQUIDS (see van Duzer [12] for a more detailed discussion of weak-link rings and SQUIDS).

When $\beta \gg 1$, for any value of applied magnetic field many metastable flux states of the ring can exist separated by φ_0 . The Birmingham observation [3] of transitions between such states within a hollow ring of multi-phase YBCO material confirmed the value of the flux quantum in HTC superconductors as $h/2e$, implying the pairing of electrons as in conventional BCS superconductors. This experiment also demonstrated the existence of long-range order of the superconducting order parameter around a path involving many thousands of superconducting grains weakly coupled together across grain boundaries. For the multi-phase YBCO ring on which the measurements were made, the superconducting loop appeared fortuitously to be broken by a single thermally recyclable Josephson junction—presumably the weakest point on a particular percolating superconducting path around the ring. The measurements were made at 4.2 K, where no evidence of thermal excitation between flux states was observed.

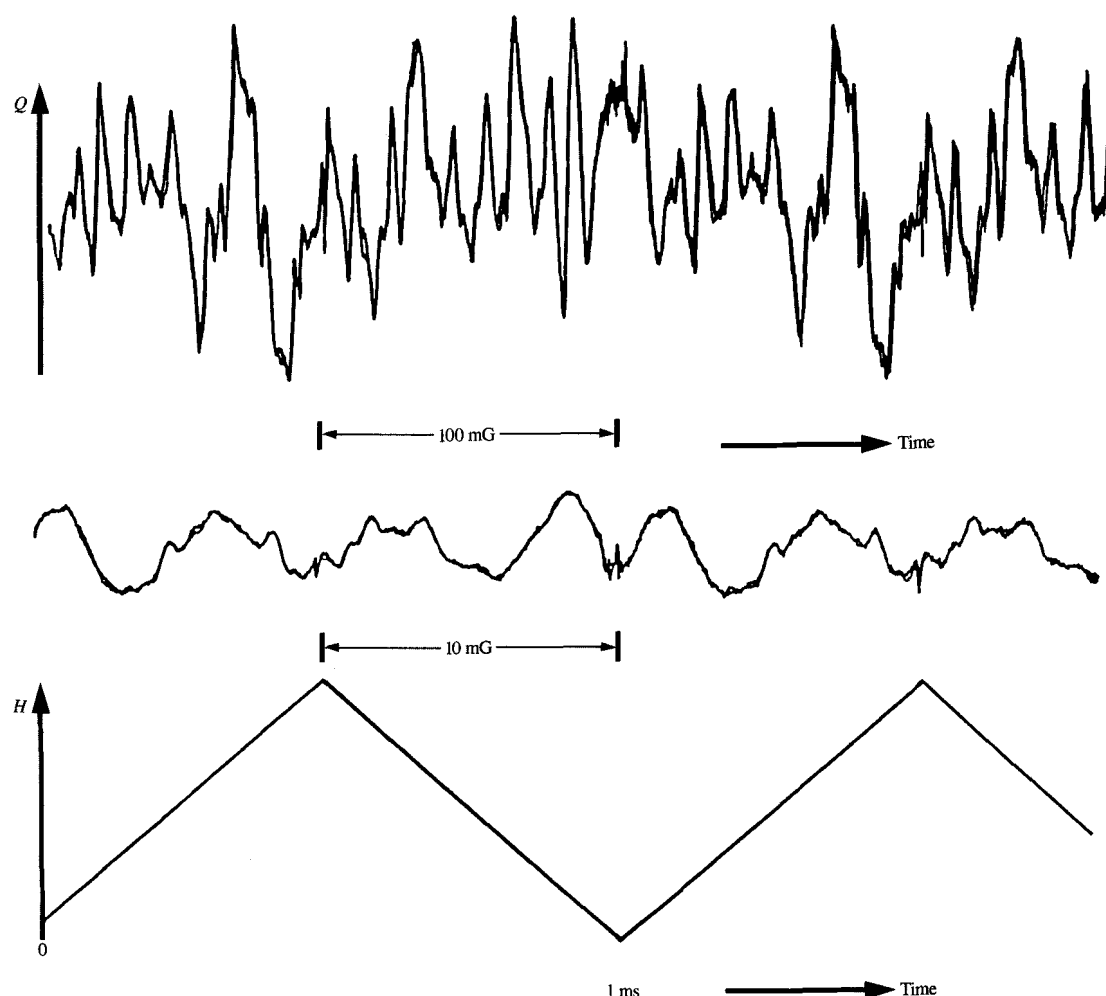


Figure 1

Quasi-field periodicity of 20-MHz rf losses in a 9-mg sample of YBCO at liquid-nitrogen temperatures as a function of linearly ramped external magnetic field for two field amplitudes.

4. Influence of Josephson tunneling on bulk properties

At low fields, a number of properties of granular ceramic HTC superconductors have been observed to vary quasi-periodically as a function of applied field—such as electrical resistance [11], rf losses (i.e., bulk rf SQUIDs) [13–16], as shown in Figure 1, and microwave losses [17–20]. These fluctuations are believed to arise from flux quantization within the superconducting loops between individual grains of the ceramic superconductor [13] and in weak-link regions within single grains [20]. The properties of the intergranular loops are expected to be periodic in the flux quantum with a

typical field periodicity $\sim \Phi_0/A^2$, where $A \sim a^2$ is the area of effective field penetration between grains. In practice, there is often appreciable field penetration into the grains, which are often comparable in size to the effective magnetic penetration length, " $\lambda(T)$." Although contributions from many different-sized loops will tend to cancel each other out, the remanent quasi-periodic field dependence will still reflect the average area of the intergranular loops.

The existence of macroscopic superconducting loops between grains may also affect the bulk electrical and magnetic properties. By analogy with a single weak-link superconducting loop, we can define an effective β value for

a granular material, $2\pi\mu_0 a i_c / \varphi_0$, where we have assumed an effective intergranular inductance L of order $\mu_0 a$. It might be expected that the magnetic properties of the individual loops, and therefore of the sample as a whole, would change from being reversible to irreversible when $\beta \sim 1$. Unless the intergranular weak links are sufficiently strong to trap flux, any superimposed current will cause flux motion and hence result in a finite resistivity.

It is a common observation that bulk superconductivity in intergranular superconductors occurs well below the onset of superconductivity in the individual grains, as illustrated in Figure 2. This shows superimposed measurements of electrical resistance and ac susceptibility for a good but not optimally processed YBCO ceramic superconductor. The onset of bulk superconductivity at T^* is indicated by the drop in electrical resistance to zero and the onset of a peak in the imaginary component of the susceptibility associated with hysteresis resulting from the trapping of flux between grains when the sample can support a supercurrent. Between T_c and T^* the measured properties are effectively reversible with respect to changes in the temperature and applied field. However, below T^* , where flux can be trapped within individual intergrain loops, the magnetic and electrical properties are hysteretic.

There are two conditions which might need to be satisfied for the occurrence of bulk superconductivity. The first essential requirement is that thermal fluctuations should not destroy the long-range coherence of the superconducting order parameter, which requires that the Josephson coupling energy between grains $i_c \varphi_0 / 2\pi \gtrsim kT$. The second is that the intergrain coupling be sufficiently strong to allow trapping of flux between grains, which requires that $i_c \varphi_0 / 2\pi \gtrsim \varphi_0^2 / 2L$, which is equivalent to $\beta \gtrsim 1$.

For a moderately well-sintered ceramic YBCO sample with a grain size $a \sim 5 \mu\text{m}$ and $J_c \sim 10^7 (1-t) \text{ A}\cdot\text{m}^{-2}$, where $t = T/T_c$, the above conditions would give a transition to long-range order when $(1-t) \sim 2\pi kT / (J_c a^2 \varphi_0) \sim 10^{-2}$, corresponding to a depression of the transition temperature for bulk superconductivity of $\sim 1 \text{ K}$. However, at this transition $\beta \sim (2\pi)^2 kT \mu_0 a / \varphi_0^2 (1-t) \sim 6 \cdot 10^{-2} < 1$, so that, despite the long-range phase coherence, it is unlikely that individual flux lines could be trapped in single loops between grains. For such a sample, one might expect a transition to bulk superconductivity at a lower temperature, when $\beta = 1$, which would occur when $(1-t) \sim 0.2$, a depression of the bulk transition temperature of YBCO by about 20 K, which is more typical of the observed depression when ceramic processing routes have not been properly optimized. Unfortunately, the above arguments are only qualitative; it is not easy to distinguish experimentally between the two possible criteria for bulk superconductivity. Both criteria suggest a strong correlation between the magnitude of the bulk supercurrent density J_c and the depression in bulk transition temperature, which is consistent with experiment.

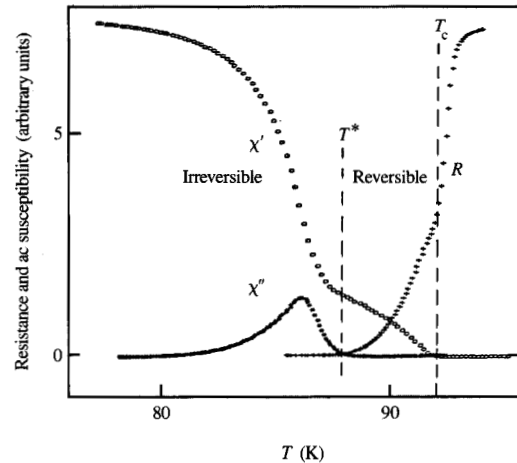


Figure 2

Temperature dependence of resistance and ac susceptibility of a typical sintered YBCO sample with modest J_c .

In an important review of the properties of granular ceramic superconductors, Clem [21] has derived the magnetic properties of granular superconductors in the limit $\beta \ll 1$. A continuum model is developed on the basis of an initial assumption of a regular array of junctions separated a distance a in a quasi-two-dimensional model. Clem has shown that the predicted magnetic properties of such a system are closely analogous to those of a conventional type-II superconductor with an effective magnetic penetration length, which may conveniently be expressed in terms of the β parameter introduced above, of order $a/\sqrt{\beta}$ and an effective coherence length $\sim a$.

The "intergranular mixed state" involves giant flux vortices with currents shared by many grains flowing over a distance equal to the magnetic penetration length, an upper critical field $H_{c2} \sim \varphi_0 / a^2$, of order $100 \mu\text{T}$ for a $5\text{-}\mu\text{m}$ intergrain size, and a very small $H_{c1} \sim \sqrt{\beta} H_{c2}$. Within this uniform continuum model, the flux lines are free to move, so that the magnetic properties would be reversible and the material resistive. In practice, local variations in pinning energies would lead to flux pinning. However, the pinning energies are likely to be rather small, so that thermally activated flux motion would result in reversible magnetic properties and a finite resistivity.

The transition between the reversible and irreversible theoretically magnetic states of ceramic superconductors has been investigated theoretically by several authors [22, 23] in terms of the superconducting glass model, first proposed by Müller, Takashige, and Bednorz [24]. In such models the

energy associated with the inductance of the intergrain current loops is usually neglected. A realistic model of a granular ceramic superconductor should take into account the three-dimensionality, the intergranular inductance energies, the inevitable variations in strengths and sizes of junctions and of void areas, and thermal fluctuations.

5. Flux creep in ceramic superconductors

A review of flux creep in HTC superconductors with references to previously published measurements has been given by Malozemoff et al. [25]. In a granular material, individual flux lines are inhibited from moving freely between the grains by the intergrain weak-link junctions, which provide a spatially varying energy barrier of height $E_0 \sim i_c \varphi_0 / 2\pi$ for motion of an individual flux line. When a supercurrent flows through the ceramic material, the energy barrier is lowered by an amount $\sim i_s \varphi_0 / 2\pi$, where i_s is the supercurrent passing through an individual junction.

Flux trapped within a hollow cylinder by virtue of circulating supercurrents will escape at a rate determined by the probability of thermally activated flux jumps between pinning sites, which is approximately proportional to

$$\omega_0 \exp\left(-\frac{E_0}{kT}\right) \exp\left(\pm \frac{\alpha i_s \varphi_0}{2\pi kT}\right),$$

where the $\pm$ signs correspond to forward and backward jumping with respect to the direction of the Lorentz force acting on the flux lines, α is a model-dependent constant of order unity, and ω_0 is a characteristic jump frequency of order 10^9 or greater. If $i_s \varphi_0 / 2\pi > kT$, which is the usual situation for the initial decay of flux, backward jumping processes can be ignored. However, at extremely long times, when the circulating currents approach zero, the difference between the energies for forward and backward jumping becomes comparable to kT , so that the backward jumping transition must also be included, leading to a slowing down of the creep rate.

Using an Anderson-Kim type of model for flux creep [26, 27] applied to an idealized model of a granular superconductor, in which flux lines are assumed to move independently by individual thermal activation across equally spaced weak-link functions (see also [28]), it can be shown that, after a very short initial transient, the field B trapped inside a thin cylinder of thickness d will decay with a logarithmic time dependence given by

$$(1/B_0) dB/d(\ln t) \sim -kT/E_0,$$

where B_0 is the initial field trapped inside the cylinder. The initial state is assumed to be described by the Bean critical-state model [29] with $dB/dr = \mu_0 J_c = \mu_0 i_c / a^2$, where a is the spacing of the weak-link junctions.

Conventional SQUID magnetometry has been used to measure flux creep out of a weakly sintered ceramic YBCO ring at very small fields of order $1 \mu\text{T}$, where one can be sure

that all the observed effects are associated with the intergranular weak links, since at such fields there can be no penetration or flux into the grains. At a given temperature, the ring is prepared in an initial critical state by lowering the external field from a relatively high value toward zero. The field is then suddenly removed so that a well-defined critical-state situation is established within the ring, with a measured value of trapped field $B_0 = \mu_0 J_c d$, where d is the thickness of the ring. The subsequent decay of trapped field is observed to be close to logarithmic at all temperatures, with a decay rate that increases approximately linearly with temperature to a broad maximum at around 35 K before decreasing again to zero on approaching T_c . Small random deviations from strictly logarithmic time dependencies were sometimes observed, which might have been associated with the release of flux trapped in deep trapping centers filled during previous thermal and magnetic cycling.

These measurements are qualitatively similar to creep measurements on granular superconductors by other authors [25, 30–32] which have usually been made at rather larger fields, where there are possible problems of interpretation because of irreversible flux penetration into the grains and subsequent creep from within the grains. Some authors report a time dependence more consistent with a Kohlrausch decay [33] reminiscent of the behavior of magnetic glasses rather than a strictly logarithmic decay. The type of decay observed may well be dependent on the type of measurement made, the time scale over which measurements are made, and sample quality.

The values for the effective E_0 deduced from the simple analysis given above were found to be very nearly constant at ~ 0.14 eV at all temperatures. This is inconsistent with the initial assumption that $E_0 \sim i_c \varphi_0 / 2\pi$, which must go to zero with $i_c(T)$ on approaching T^* (~ 65 K for the purposely weakly superconducting impurity-doped YBCO ring investigated).

One possible explanation, which has also been suggested by Malozemoff et al. [25] to account for similar observations in single crystals [34], is that, on approaching T_c , the number of junctions that can trap flux decreases, as might well be expected from our previous arguments. For example, if there is a spread in i_c values, those superconducting loops with β less than unity simply drop out of the problem, increasing the average spacing between effective trapping centers. Similarly, those junctions for which $i_c(T)$ becomes sufficiently small to allow significant backward jumping also become ineffective. Since $B_0 = \mu_0 J_c d = \mu_0 i_c d / a^2$ (the intergranular H_{c1} is assumed to be effectively zero for ceramic superconductors), and we assume that $E_0 = \varphi_0 i_c / \pi$, the expression for the logarithmic flux creep rate can be rearranged to give the average spacing of effective pinning centers,

$$\langle a^2 \rangle = -\frac{kT\pi\mu_0 d}{\varphi_0 [dB/d(\ln t)]}.$$

Assuming that this simple model is at least qualitatively correct, our measurements imply a spacing between effective pinning sites in granular material that can become very long indeed [$a \sim 30/(1 - T/T^*) \mu\text{m}$], many times the intergranular spacing on approaching T_c . Details of these measurements and a more refined analysis will be presented elsewhere [34].

6. SQUID magnetometry using ceramic superconductors

Under laboratory conditions a liquid-helium-cooled SQUID can have a sensitivity approaching the quantum limit,

$$\frac{\langle \varphi_{\text{noise}}^2 \rangle}{2L} \sim h$$

(see the recent review by Clarke [35]), and practical SQUIDs can have a sensitivity orders of magnitude better than the best available flux-gate magnetometer [36]. There is, therefore, considerable interest in developing SQUIDs which operate at LN_2 temperatures using ceramic materials.

Evidence for SQUID behavior arising from rf coupling to weak-link intergranular current loops between individual grains was first reported by the Birmingham group in their original determination of the flux quantum in an HTC ceramic ring [3]. The quasi-periodic rf SQUID field dependence observed, similar to that illustrated in Figure 1, formed the basis of the first practical magnetometer based on an HTC superconductor [13]. The scale of the observed quasi-periodicity of the field dependence suggested SQUID action in intergranular loops of typical 10–50- μm size. However, the sensitivity of magnetometers based on intergranular current loops is relatively low because of the small effective area, the field sensitivity being proportional to $\phi_0/(\text{area of loop})$.

Conventional liquid-helium-cooled niobium rf SQUIDs usually involve superconducting loops of mm size. The first such device incorporating a YBCO ceramic superconductor was developed at NBS by Zimmerman et al. [36]. This device was formed from a YBCO disc incorporating a single 1-mm hole with a weak link formed by cracking the ceramic material under liquid helium or nitrogen, creating what is known as a crack junction. Despite such a device being potentially unstable and not allowing thermal cycling with any reliability, an ideally periodic field dependence, corresponding to quantized flux in the 1-mm hole, was observed at LN_2 temperature. A flux sensitivity of order $5 \times 10^{-4} \phi_0/\sqrt{\text{Hz}}$ was reported at LN_2 , only a few times worse than typical noise figures for commercially available liquid-helium-cooled rf SQUIDs [37].

Subsequently Zavaritski et al. [38] in Moscow and Harrop et al. [39] in Birmingham developed monolithic two-hole versions of such a device, with the Josephson weak link formed by a narrow bridge between the two holes, as illustrated by the inset in Figure 3. Figure 3 shows the

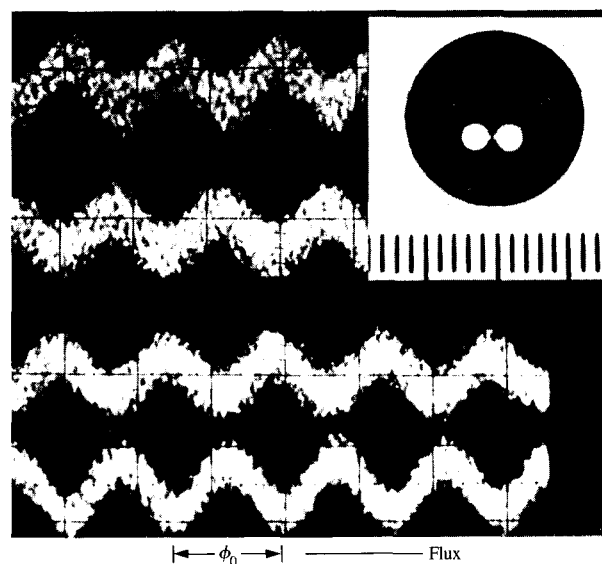


Figure 3

Quantum field periodicity of rf losses in a bulk two-hole rf SQUID (shown in inset) at liquid-nitrogen temperatures.

periodic field dependence of a Birmingham SQUID, carefully fabricated from a high-quality, single-phase YBCO disc, illustrating the predicted phase reversals on increasing the rf amplitude level [12]. When flux was applied by a long solenoid threading one of the holes, ideal field periodicity was observed for many hundreds of flux quanta. This permitted a measurement of the flux quantum in YBCO at liquid-nitrogen temperatures to be made to an accuracy limited only by the uncertainty (probably $\sim 5\%$), in determining the exact current paths in the two holes [39], (an absolute determination requires the relative sizes of the two holes to be accurately known). Unlike the field dependence observed in granular rf SQUIDs, the observed periodicity for the monolithic rf SQUID is ideally reversible for applied flux changes of many hundreds of flux quanta.

For many applications, the ultimate performance of a SQUID is determined by its low-frequency noise. The noise spectrum of a typical Birmingham two-hole SQUID at LN_2 temperatures is shown in Figure 4, illustrating the dominance of an approximately $1/f$ source of noise at low frequencies (see also [40]). All groups working on both bulk and thin-film SQUIDs observe $1/f$ noise of comparable values, and such noise may be intrinsic to granular HTC materials. A likely cause is flux noise generated by thermally activated flux motion of trapped flux, which is clearly closely related to the flux-creep phenomenon. Above 100 Hz the sensitivity is largely limited by the room-temperature electronics. Improvements in the performance will undoubtedly be obtained by the use of LN_2 -cooled amplifiers

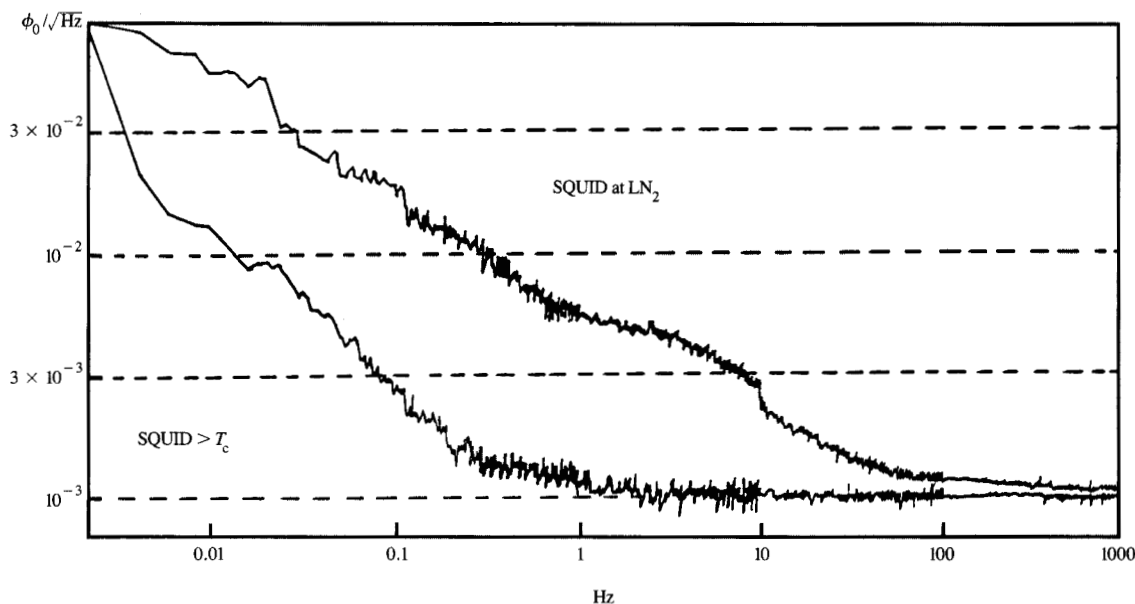


Figure 4

Noise measurements for a two-hole rf SQUID obtained at 77 K and above T_c to distinguish the contribution from electronic noise alone.

and superconducting tank circuits and by optimized coupling. However, these measurements already demonstrate the viability of the LN_2 rf SQUID as a potential competitor to conventional magnetometers and even to liquid-helium-cooled conventional SQUIDs in situations where environmental noise is likely to be a significant factor affecting achievable sensitivity.

7. dc SQUIDs using ceramic materials

dc SQUID operation is based on the periodic field variations of the voltage measured across a superconducting loop containing a matched pair of resistively damped junctions in parallel [12]. A very early realization of an HTC dc SQUID was reported by de Waele et al. [41], who observed almost ideal flux quantum oscillations of the voltage across two pieces of ceramic material, which were simply pushed together. On increasing temperature, quasi-periodic variations were observed up to 68 K. More recently Robbes et al. [11] observed similar oscillations in a ceramic superconductor that was purposely thinned down to give a narrow constriction region with the current confined to a relatively small number of grains. In both experiments, the observed quasi-periodicity corresponded to an effective area comparable to that expected between grains. As in granular rf SQUIDs, the sensitivity of such devices is limited by the

relatively small size of the intergranular current loops involved.

There have been a number of attempts to devise dc SQUIDs using thin films of YBCO and other HTC superconductors. The earliest such SQUID was reported by Koch et al. [42], who lithographically patterned a 40- μm -square central hole and two 10- μm weak-link regions in a thin polycrystalline YBCO film of 5 μm typical grain size. The strength of the Josephson tunneling in the weak-link regions was adjusted by ion implantation. At liquid-helium temperatures the I/V characteristics exhibited the anticipated field periodicity, but when the temperature was increased the field dependence became increasingly aperiodic, reminiscent of the field dependences observed for current loops, presumably between the grains of the polycrystalline thin films. All evidence of SQUID operation again disappeared at 68 K, almost certainly because aging and lithographic processing reduced the critical temperature of the film.

Similar devices have been reported by other groups [10, 43, 44] but it has proved difficult to establish satisfactory LN_2 -temperature dc SQUID operation using YBCO thin films. However, Koch and co-workers [45] at IBM Yorktown Heights have recently reported a TI-based thin-film SQUID with a very encouraging noise performance at liquid-nitrogen temperatures, which is already comparable to

a commercial liquid-helium rf SQUID. However, $1/f$ noise remains a problem, though to a lesser extent than in any previously reported HTC SQUID.

The sensitivity of a SQUID will always be limited by thermal noise [35, 46], so that LN_2 SQUID operation will inevitably be intrinsically less sensitive than liquid-helium operation. However, for many applications where the ultimate in sensitivity is not required, the convenience of LN_2 operation may well outweigh the slight loss in sensitivity with increased temperature.

Acknowledgments

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