

## Accurate Determination of $2e/h$ in Y-Ba-Cu-O Josephson Junctions

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Accurate measurements of  $2e/h$  by means of the ac Josephson effect have been carried out by the use of weak links of a high- $T_c$  superconductor, Y-Ba-Cu-O, of macroscopic dimensions. The result,  $2e/h = 483\,595.0 \pm 1.7$  GHz/V, differs, in relative value, from that obtained by accurate measurements in metallic superconductors by  $5.6 \times 10^{-6}$ .

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Much experimental effort is now directed toward the characterization of superconductivity in the high- $T_c$  compounds<sup>1</sup> to determine which phenomena are common to all known superconductors and which are particular to the high- $T_c$  compounds. Josephson's theoretical treatment<sup>2</sup> of two weakly coupled superconductors led to a series of effects. The ac Josephson effect refers to the flow of alternating current between two weakly coupled superconductors when a potential difference  $U$  is maintained between them. The frequency of this current is

$$f = (2e/h)U, \quad (1)$$

where  $e$  is the elementary charge and  $h$  is Planck's constant. Equation (1) follows from very general assumptions about superconductors.<sup>3</sup> Various tests of the dependence of Eq. (1) on experimental parameters have been carried out in metallic superconductors.<sup>4</sup> The accuracy of Eq. (1) is so high that after being used to determine  $2e/h$ , it is now the basis for primary reference standards of voltage in national standards laboratories.

The observations of flux quantization in units of  $h/2e$  in bulk rings<sup>5</sup> of Y-Ba-Cu-O and of microwave-induced voltage steps in weakly linked samples<sup>6,7</sup> motivated this attempt to accurately verify Eq. (1) in the same material. This Letter reports the first highly accurate determination of  $2e/h$  in a high- $T_c$  material.

Y-Ba-Cu-O samples were prepared by the solid-state reaction method.<sup>8</sup> Following the third calcination, bars of dimensions  $15 \times 1.8 \times h$  mm<sup>3</sup> ( $0.5 \leq h \leq 2$ ) were pressed, sintered, and made to react with oxygen. Gold contacts and indium solder were used to form three pairs of contacts to the sample. From dc-resistance measurements, the bulk  $T_c$  was found to be 92 K and the transition width (10% to 90%) 1.4 K. The bars were cemented to Corning 7059 glass substrates of dimensions  $25.4 \times 25.4 \times 1.2$  mm<sup>3</sup>. The constriction forming the weak-link Josephson junction was filed or sawed by hand under a microscope. A typical cross-sectional area of the constriction is  $0.1$  mm<sup>2</sup>.

The current-voltage characteristics of these structures are those of resistively shunted Josephson junctions.<sup>9</sup> The response to electromagnetic radiation is broadband and the key element in producing constant-voltage steps

is to rigorously avoid, by filtering and screening, any spurious radiation. Filtering was conveniently done by mounting samples in a microwave integrated-circuit box<sup>10</sup> fitted with low-pass filters on all dc leads. In addition, all leads into the cryostat were carefully filtered with two or three stages of  $\pi$ -type filters. The microwave radiation source is a phase-locked, X-band klystron having a relative frequency stability of about  $1 \times 10^{-9}$ /min and a narrow bandwidth. Inside the cryostat the transmission line is 3.6-mm semirigid cable. All measurements were carried out in metal helium or nitrogen storage Dewars. All of the equipment was operated inside a screened room.

The complete experimental arrangement is indicated schematically in Fig. 1. The bias current supply produces either alternating current for preliminary adjustments of steps or direct current for biasing on the step to be measured. Variable-offset voltage sources, not shown, allowed the visualization of steps on the oscilloscope at voltage and current resolutions of tenths of microvolts and tenths of microamperes.

The output voltage of the junction was measured in terms of an accurately known standard cell with a modified Julie PVP 1001J potentiometer and model VDR 307J Kelvin-Varley divider.<sup>11</sup> For this work, the most important feature of this potentiometer is that, in

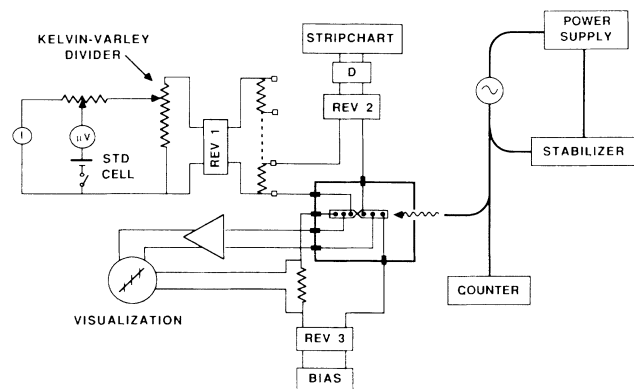


FIG. 1. Schematic diagram of the experimental arrangement.

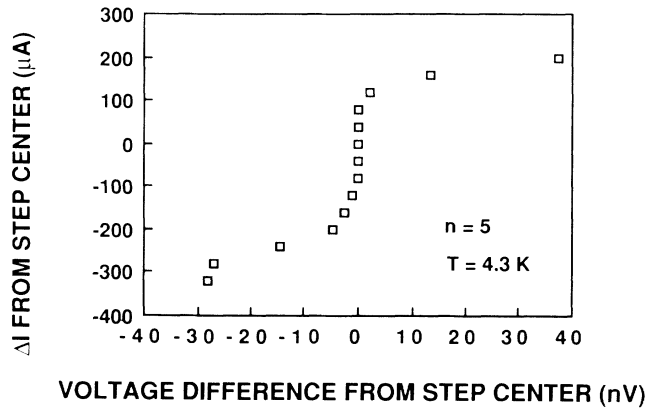


FIG. 2. High-resolution plot of the shape of a step. The size of the squares approximates the resolution in current and voltage.

addition to having been calibrated by a tedious classical technique,<sup>11</sup> it was also calibrated on the 1-mV range directly against the output of the Pb-Pb<sub>x</sub>O<sub>y</sub>-Pb tunnel junctions which have been used to realize<sup>12</sup> the Bureau International des Poids et Mesures voltage standard (denoted by  $V_{76-BI}$ ). The uncertainty in the value of the standard cell in Fig. 1 in terms of  $V_{76-BI}$  is of the order of 100 nV. The detector  $D$  is an EM Laboratory model N1A nanovoltmeter fitted with an isolated output to drive a pen recorder, allowing integration of the voltage balances and giving permanent records of the measurements. The measurement procedure was such that the effects of thermal emfs and their linear drifts in the measuring circuit and the junction leads were eliminated by polarity reversals.

It is known that external voltage noise can greatly increase the dynamic resistance of a step induced by radiation and that external current noise will reduce the range of current over which the step voltage remains constant.<sup>13</sup> Furthermore, it is also well established that thermal noise can produce significant dynamic resistance

of a step.<sup>14,15</sup> It was, therefore, of crucial importance to carefully check the step profile while setting up a measurement run and to recheck the setting of the bias current, by sweeping over a small range of the step with the highest voltage sensitivity, every time the bias current is reset. Figure 2 shows the result of a detailed sweep of a step made by manually stepping the bias current.

Table I lists the experimental conditions, uncorrected results, and the critical potentiometer corrections with uncertainties (all uncertainties are intended to be either measured standard deviations or  $1\sigma$  estimates). Table II lists the random uncertainties of the same runs illustrating the decreased random uncertainty of the last three runs due to the technique of accurate verification of the absence of resistance of the steps for each measured point. The uncertainty due to potentiometer calibration is reduced for the last three points because, for them, the potentiometer was calibrated by direct comparison with the output of Pb-Pb<sub>x</sub>O<sub>y</sub>-Pb tunnel junctions. The remaining noteworthy uncertainties are due to variations of potentiometer supply current and thermal emfs in the instrument.

The total root-sum-square uncertainties in column 5 of Table II were used to calculate a weighted mean and standard deviation of the results. The result is

$$2e/h |_{Y-Ba-Cu-O} = 483\,591.3 \pm 1.7 \text{ GHz}/V_{76-BI}. \quad (2)$$

In comparison with the uncertainty given above, possible systematic effects which would not vary from run to run were estimated to be negligible, mainly because the potentiometer was calibrated under the same conditions as those in which it was used. One possible exception to this assertion is that, in this work, the oscilloscope with its 1 M $\Omega$  resistance to ground may have introduced leakage paths in a circuit normally operating at about  $10^{11}$   $\Omega$  above ground.

The representation of the volt at Bureau International

TABLE I. Summary of experimental conditions.

Run No.	Sample	Temperature (K)	$n$	Approximation $U$ ( $\mu$ V)	Uncorrected $2e/h-483\,000$ (GHz/ $V_{76-BI}$ )	Correction to $U$ and uncertainty (nV)
1	6/2	77	1	21.23	590.06	$0 \pm 1$
2	6/4	$\sim 8$	6	143.93	577.38	$2.5 \pm 1$
3	6/4	$\sim 8$	5	119.93	612.58	$2 \pm 1$
4	6/2	4.2	4	82.50	555.89	$0 \pm 1$
5	6/2	4.2	5	110.13	596.58	$2 \pm 1$
6	6/2	4.3	5	119.61	581.69	$2 \pm 1$
7	6/2	$\sim 8$	4	96.41	599.12	$0.6 \pm 1$
8	6/11	4.2	8	194.33	599.83	$3.1 \pm 0.3$
9	6/11	4.2	13	315.77	596.51	$2.1 \pm 0.3$
10	6/11	4.2	14	340.06	594.93	$3.0 \pm 0.3$

TABLE II. Summary of experimental results and uncertainties.

Run No.	No. obs.	Standard deviation of the mean (GHz/V)	Corrected $2e/h-483000$ (GHz/V <sub>76-BI</sub> )	Total variable component of uncertainty (GHz/V)
1	4	24.18	590.06	46.27
2	2	3.53	568.81	6.81
3	5	3.05	604.52	7.62
4	5	18.68	555.89	21.42
5	2	12.57	587.80	14.69
6	9	9.67	573.60	11.94
7	3	16.44	596.11	18.60
8	10	1.60	592.05	3.29
9	3	0.35	593.33	1.81
10	6	0.58	590.66	1.75
Weighted mean and standard deviation			591.3	1.7

des Poids et Mesures is defined in terms of a conventional value of the Josephson frequency/voltage quotient namely of  $483594.0 \text{ GHz/V}_{76\text{-BI}}$ . The latest (1986) CODATA adjustment of the physical constants<sup>16</sup> gives  $V_{76\text{-BI}} = [1 - (7.59 \pm 0.30) \times 10^{-6}] \text{ V}$ , so that

$$2e/h|_{\text{Y-Ba-Cu-O}} = (483595.0 \pm 1.7) \text{ GHz/V}. \quad (3)$$

This differs from the 1986 CODATA value of  $483597.67 \pm 0.14 \text{ GHz/V}$  by  $2.7 \text{ GHz/V}$  or about 1.6 times the estimated standard deviation. The results of this study are thus in good agreement with those obtained in metallic superconductors.

The results of Table I were obtained on integral-valued steps,  $n$ . Some of the junctions also exhibited subharmonic steps of order  $n = 5\frac{1}{2}$  similar to those observed in point-contact or metallic-microbridge junctions. Although this step was not measured with the potentiometer, a digital nanovoltmeter was used to verify that it implied a value of  $2e/h$  of  $(483860 \pm 100) \text{ GHz/V}$ , in good agreement, considering the uncertainty, with those of the more carefully measured integral-valued steps.

The constriction junctions used in this work have microwave<sup>7</sup> and dc<sup>17,18</sup> properties similar to resistively shunted Josephson junctions microbridges. Since microbridge behavior occurs in metallic superconductors only if the bridge dimensions are of the order of magnitude of the coherence length and since the coherence length in Y-Ba-Cu-O is of the order of 1.2 nm,<sup>19</sup> the mechanism responsible for the junction behavior is not well understood. Recent observations<sup>18</sup> of the Josephson coupling, inside and at the boundaries of grains in Y-Ba-Cu-O films suggest that the constriction junctions used in this work may be three-dimensional networks of junctions. The fact that these structures are capable of yielding an accurate value of  $2e/h$  strongly suggests that they are bona fide Josephson junctions.

In conclusion, an accurate value of  $2e/h$  has been obtained from ac Josephson-effect measurements in constriction junctions in Y-Ba-Cu-O and it nearly agrees, to within the estimated uncertainty, with that obtained in metallic superconductors. This strongly implies that these macroscopic structures are indeed Josephson junctions.

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