EXPERIMENTAL COMPARISON OF THE JOSEPHSON VOLTAGE-FREQUENCY RELATION IN DIFFERENT SUPERCONDUCTORS*

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Using the ac Josephson effect we have demonstrated experimentally that $2e/h$ is identical in lead, tin, and indium to within 1 part in 10^8.

Josephson[1] showed that when two weakly coupled superconductors are biased at a voltage V, an alternating supercurrent of frequency

$$\nu = 2eV/h \tag{1}$$

flows between them, e being the electronic charge and h Planck's constant. The effect of electromagnetic radiation of frequency ν upon a Josephson junction or weak link is to induce time-independent constant-voltage current steps[2] on the current-voltage characteristic. In the case of a pure Josephson junction in which the current through the barrier is related to the difference in the phase of the order parameter (φ) across it by the relation[1] $j = j_1 \sin\varphi$, the voltage at which the steps appear is given by

$$V_n = nh\nu/2e, \tag{2}$$

n (the order of the step) being an integer. In the more general case of a metallic bridge or point contact, subharmonic steps may also be observed.

By irradiating junctions at a known frequency and measuring the voltages at which the resultant steps occur, Parker, Taylor, and Langenberg[4,5] have deduced an absolute value of e/h with a precision of 6 parts in 10^6. This result, provided it contains no unsuspected systematic error, resolves a major difficulty in modern quantum electrodynamics, namely the discrepancy between the theoretical and experimental values of the hyperfine splitting of atomic hydrogen. In addition, the method provides a simple and accurate means of comparing and maintaining standards of electromotive force.[6] Consequently it is of importance to establish Eq. (2) as an exact universal relation which is independent of the superconductors used and the conditions under which the experiment is performed. Parker, Taylor, and Langenberg showed that this was so to within 2 parts in 10^6. The present experiments, although of low absolute accuracy, demonstrate that the voltage-frequency relation is identical in different materials to 1 part in 10^8. The principle of the experiment is to apply the same radiation to two junctions of dissimilar materials and bias each on the induced step of the same order n: By directly comparing the voltages across the two junctions, one may make a very precise differential measurement.

In contrast to the insulating barriers or point contacts used in previous experiments, the present junctions consisted of a normal-metal barrier sandwiched between two superconductors.[7] The junctions were made by evaporating successively onto a glass substrate a strip of superconductor (lead, tin, or indium), a disk of normal metal (usually copper), and finally a second strip of superconductor at right angles to the first [see Fig. 1(a)]. The superconductors were about 0.2 mm wide and 5000 Å thick; the copper was typically 10 000 Å thick. The current-voltage characteristics of these sandwiches were studied in the four-terminal arrangement shown. It was observed that the junctions sustained a supercurrent up to a certain critical value, above which a voltage appeared, rising smoothly to a linear dependence on current. Because of the very low electrical resistance of the copper barrier, typically 10^{-7} Ω, the voltages developed

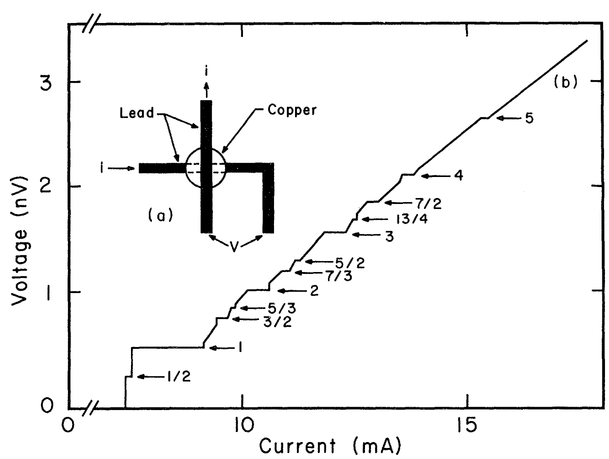

FIG. 1. (a) Lead-copper-lead junction configuration. (b) The current-voltage characteristic of a lead-copper-lead junction at 4.2°K irradiated at a frequency of 250 kHz.

were small and it was convenient to use a superconducting voltmeter[8] to measure them. The critical current, typically a few milliamperes, decreased exponentially with increased thickness of copper and rose rapidly as the temperature was lowered.[7] The magnetic field dependence of the critical current indicated that the sandwiches behaved essentially as Josephson junctions.

A sinusoidal rf field was applied to the junctions by means of an oscillator driving a small solenoid surrounding them. Current steps were induced on the characteristic; thus both ac and dc Josephson effects are observed in these sandwiches. Steps were seen at frequencies between a few kilohertz and about 1 MHz, the latter corresponding to a voltage of about 2.1 nV at the first harmonic step. The amplitude of the steps was rapidly attenuated at voltages higher than a few nanovolts, irrespective of the frequency of the applied radiation. Thus with a frequency of 1 MHz only the first harmonic step was visible, whereas at 100 kHz steps were observed up to at least the tenth harmonic. An example of the induced steps is shown in Fig. 1(b). Subharmonic steps were also usually observed, as in the thin film bridges studied by Anderson and Dayem,[3] indicating that flux flow occurs in the junctions at finite voltages. It is not yet known whether the true ac Josephson effect or a flux-flow mechanism is dominant in producing the integer steps. However, the distinction is immaterial for the present purpose since in both processes the same quantity $2e/h$ relates voltage and frequency. The important feature which makes possible the very precise voltage measurements is the extremely low differential resistance of the steps compared with that of other types of junction.

The differential resistance of the steps was measured in the following manner. Two junctions, of identical superconductors, were connected in series with a superconducting galvanometer[8] and a superconducting switch (see Fig. 2). The resolution of the galvanometer was 0.3 μA. The superconducting thermal switch was tested with subcritical currents in the junctions and did not induce any observable spurious currents into the circuit. (The circulating current induced due to flux quantization was no greater than $\frac{1}{2}\varphi_0/L \simeq 10$ nA.) The entire circuit was surrounded by a superconducting can to screen out external magnetic field fluctuations and immersed in a bath of superfluid helium, the temperature of which was stabilized to within 10^{-4}°K. The cryostat was surrounded by a double Mumetal

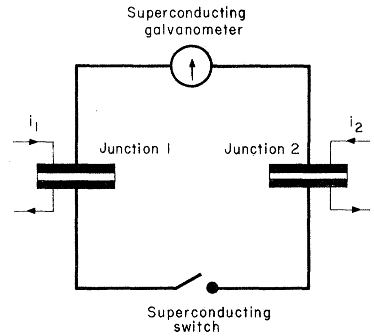

FIG 2. Circuit used both for measuring the differential resistance of the induced steps and for comparing the voltages developed across two junctions made from different superconductors. The currents i_1 and i_2 are adjusted to bias each junction on the same order current step.

shield to reduce the earth's field to a few milligauss. The inductance of the circuit (L), estimated by introducing a known resistance in series with the galvanometer and measuring the time constant, was $(1.0 \pm 0.1)\times 10^{-7}$ H. The junctions were subjected to the same radiation and with the switch open each was biased independently on the same-order induced step. When the switch was closed, no detectable current flowed through the galvanometer and each junction remained biased upon its step. In this state, although there was a voltage across the junctions, the circuit was superconducting in the sense that a magnetic field applied to it induced a circulating supercurrent, provided that its amplitude was insufficient to drive either junction off the induced step. In order to obtain the maximum possible circulating current, junction 1 was current-biased at the lower end of its step and junction 2 at the upper end, and the circulating current established in the same direction as i_1. An upper limit was set on the differential resistance of the steps by studying the decay of the circulating current.

In a typical experiment, the frequency of the applied radiation was 500 kHz, so that the voltage of the first-order induced step, upon which each junction was biased, was approximately 1.06 nV. The amplitude of the step was typically 0.5-1.0 mA. With an induced circulating current of 500 μA, the decay was less than 0.3 μA in 1.8×10^3 sec, giving a time constant (τ) greater than 3×10^6 sec. Thus the total circuit resistance (L/τ) was less than 3.3×10^{-14} Ω. We see that the voltage width of a step with an amplitude of 0.5 mA was less than 10^{-17} V, a value many orders of magnitude smaller than that obtained with point contacts or insulating barriers.

It was found that exactly the same results were obtained when the junctions were of dissimilar superconductors. Further measurements were then made to set an upper limit on the voltage difference between the two junctions. In the initial experiments the junctions were lead-copper-lead and tin-copper-tin: This combination was chosen because lead is a strong-coupling superconductor whereas tin is not. If there were a voltage difference (ΔV) between the junctions, then a circulating current (j) would be set up at an initial rate $dj/dt = \Delta V/L$. When the superconducting switch had been closed for 1.8×10^3 sec, the circulating current was still less than the galvanometer resolution (0.3 μA) so that ΔV was no greater than 1.7×10^{-17} V. These experiments were performed at frequencies up to 1 MHz at which frequency the voltage of the first-order induced step was about 2.1×10^{-9} V. We may therefore say that the voltages developed across the junctions differed by less than 1 part in 10^8.

The same null result was obtained when the pairs of superconductors were lead and indium or tin and indium and also when silver was substituted for copper as the barrier in one of the junctions. In addition, the variation of the following parameters, although generally affecting the shape of the current-voltage characteristic and the amplitude of the steps, did not give rise to any observable difference in the voltage across the junctions: (i) temperature, from 1.2°K to 2.2°K; (ii) the thickness of the barrier; (iii) the level of the applied rf power, over a factor of 5; (iv) the rf frequency, from 100 kHz to 1 MHz (to 1 part in 10^7 at 100 kHz); (v) the order of the steps on which both junctions were biased, up to the fourth order; (vi) the position on the induced step; (vii) the ambient magnetic field, up to ±1 G; (viii) the direction of the bias current through the junctions.

During the course of each measurement, both the frequency and amplitude of the rf drifted by about 1 part in 10^5. The frequency shift caused the same change in the voltage induced across each junction and the amplitude variation affected the shape of the current-voltage characteristic and the size of the steps. However, neither fluctuation was sufficiently large to drive the junctions off the steps or to introduce any discernable dissipation of the circulating current.

In conclusion we may state that $2e/h$ is identical in lead, tin, and idium to within 1 part in 10^8 under a variety of experimental conditions. From these observations we cannot deduce directly that the charge on the pairs[9] has the free-electron value to within these limits. However, it would be truly remarkable if large corrections to the free-electron charge exist which are identical for all three metals at this level of precision. It is more likely that any corrections would be on the order of, or less than, 1 part in 10^8. One might, therefore, express considerable confidence in the absolute accuracy of the results of Parker, Taylor, and Langenberg at the level of parts in 10^6, as regards both the quantum electrodynamic implications and the proposal for comparing and maintaining standards of voltage.

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[7]A detailed account of the preparation and properties of these junctions is to be published.

[8]The superconducting galvanometer used in all of the experiments has been described elsewhere [J. Clarke, Phil. Mag. 13, 115 (1966)]. The galvanometer consists of a pellet of tin-lead solder surrounding a length of niobium wire so as to form a weak-link device. The critical current is a function of the current flowing in the wire. To avoid feeding spurious ac signals into the cryostat, a dc technique [J. W. McWane, J. E. Neighbor, and R. S. Newbower, Rev. Sci. Instr. 37, 1602 (1966)] was used to monitor the critical current and hence detect changes in the current flowing along the niobium wire. In the determination of the characteristics of the Pb-Cu-Pb junctions, a resistance of about 10^{-7} Ω was connected in series with the galvanometer and the voltages measured using a null-reading technique.

[9]In his thesis, Parker points out that if higher order

correlations of electrons are present (e.g., 4 or 6) these will give rise to subharmonic or lower order harmonic steps and not affect the voltage at which the steps appear.

REFLECTANCE MODULATION BY THE SURFACE FIELD IN GaAs

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By studying reflectance spectra in GaAs modulated by a second, intense light beam, we have observed oscillatory structures in the spectra near the energy gap as well as near 3 eV. We conclude that these structures result from a neutralization of the built-in surface field by free carriers created by the intense light beam. The experimental line shape near the band gap is qualitatively very similar to the theoretical prediction for the Franz-Keldysh effect but is shifted to lower energies presumably due to exciton effects.

We report photoreflectance (PR) experiments at 77° and 294°K in GaAs in which the reflectivity is modulated by a second, intense light beam. PR experiments were first reported by Wang, Albers, and Bleil[1] for several materials at room temperature. These authors suggested that the reflectance is modulated by the built-in surface field which is periodically neutralized by the free carriers created by the intense light beam. Gay and Klauder[2] have recently proposed that the PR effect observed by Wang, Albers, and Bleil[1] is due to a change in the effective density of states produced by the photoexcited carriers via the Pauli principle, i.e., band filling. On the other hand, if the modulation is in fact produced by an electric field, its origin may be either the Dember field which arises from the difference in mobility of the photoexcited electrons and holes or the neutralization of the surface field. The results reported here allow us to identify the mechanism responsible for photoreflectance in GaAs.

As the source of the modulating light we have used a He-Ne laser at 6328 Å to facilitate studies of the dependence of the PR line shape upon the intensity of the modulating beam. The various mechanisms proposed to explain the PR effect differ in the predicted dependence of the line shape upon the intensity of the modulating beam. Both light beams were near normal incidence on the samples and the laser was chopped at 510 Hz. Our experiments were performed on as-grown surfaces of high-purity epitaxial layers grown by K. L. Lawley at Bell Telephone Laboratories. Both n-type and p-type samples with impurity concentrations in the range 10^{15} to 10^{16} cm^{-3} were used. For the experiments at 77°K the sample was immersed in liquid nitrogen. Assuming nucleate boiling of the liquid nitrogen[3] the average laser power of 0.150 W/cm^2 raised the temperature of the sample at most ~2°K above the temperature of the bath. The periodic temperature variation at 510 Hz was, of course, much smaller. From the known thermal diffusivity[4] of GaAs we estimate[5] that the peak-to-peak temperature swing at 510 Hz was ~0.015°K.

In Fig. 1 we present the PR spectrum for photon energies near the band gap in GaAs for both 77 and 294°K. At both temperatures, six peaks are resolved with the spacing of the peaks being somewhat smaller at 77°K than at 294°K. In addition, a very sharp spike labeled B in Fig. 1 appears at 77°K. Apart from this spike, which will be discussed in a later section, the PR line shape is qualitatively very similar to the theoret-

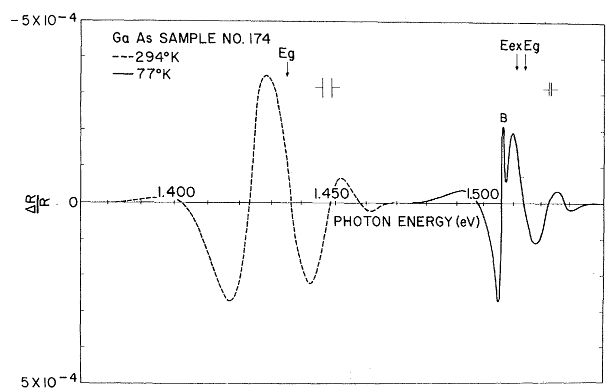

FIG. 1. Photoreflectance spectrum for photon energies near the band gap of GaAs, n-type epitaxial layer, $n \approx 10^{15}$ cm^{-3}. E_g and E_{ex} indicate the energies of the band gap and free exciton as determined by Sturge [Phys. Rev. 127, 768 (1962)]. The peak signal-to-noise ratio is ~100.