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Comparison of Microwave-Induced Constant-Voltage Steps in Pb and Sn Josephson Junctions*

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The microwave-induced voltage steps at $V = nh\nu/2e$ in the current-voltage characteristics of lead and tin Josephson junctions have been compared using a superconducting-quantum-interference device (SQUID) as a null detector in a potentiometer with 2×10^{-13}-V resolution. The junctions are individually biased and then connected to the voltmeter by means of a superconducting mechanical switch. The incident microwave power is provided by two phase-locked x-band klystrons whose variable difference frequency is stable to better than 1 part in 10^{12}. The upper limit on the relative voltage difference between the $n = 15$ (300-μV) voltage steps for a Pb-PbO and Sn-SnO junction is $\Delta V/V \leq 5\times10^{-9}$. An apparent systematic error associated with switching prevents further absolute accuracy.

INTRODUCTION

Recently, there has been much discussion of the limits of validity of the Josephson voltage-frequency relationship $h\nu = 2eV$.[1-3] The theoretical expectations for the relative precision of this expression far exceed present experimental limits, which therefore merit further extension. Experiments by Clarke[4] using superconductor-normal-superconductor (SNS) junctions have indicated that to 1 part in 10^8 the frequency-voltage relationship is identical in different materials. The experiment reported here is a similar differential test of the Josephson relationship. It entails a differential comparison of the microwave-induced constant-voltage steps of two different thin-film junctions at the level of 1 part in 10^9. The voltage difference between identical steps on the current-voltage characteristics of the two junctions is measured with a superconducting-quantum-interference device (SQUID)[5,6] in a potentiometer mode with a resolution of better than 2×10^{-13} V. Initial measurements have been made with a Pb and Sn junction simultaneously biased on the $n = 15$ constant-voltage step when irradiated with microwaves at 9.870 GHz.

A conceptual representation of the experiment is shown in Fig. 1. Two Josephson junctions J_a and J_b are irradiated with x-band microwaves at frequencies ν_a and ν_b, and biased independently on the same constant-voltage step. The voltages across the two are then $V_a = nh\nu_a/2e$ and $V_b = nh\nu_b/2e$, where n is the order of the step. A mechanical superconducting switch is then closed and any voltage difference $\Delta V = V_a - V_b$ appears across a series resistor R_c. This voltage is detected by the voltmeter and is recorded as the control voltage necessary to null the potentiometer. Although a current $i = \Delta V/R_c$ can flow when the switch is closed, this is not large enough to pull the junction off the constant-voltage step even with frequency differences as large as 10 kHz. The difference frequency ΔV is stabilized to better than 1 part in 10^{12}. The limit of the comparison is determined by the resolution of the voltmeter and the noise introduced in switching. This latter contribution sets the limit in the present data.

The feature of this experiment which allows the high precision is the mechanical superconducting switch with a completely open off state. The high (infinite) resistance of the open switch permits the use of relatively-high-resistance tunnel junctions which exhibit steps at large voltages $V > 100$ μV. The inclusion of a normal resistor in the measuring circuit eliminates any quantum coherence between the two junctions. The use of separate phase-locked microwave sources provides the capability to demonstrate a true null by varying the frequency of radiation incident on one of the junctions.

APPARATUS

The experiment is performed in a glass-walled helium Dewar at a temperature of about 2.1 °K, which is stable to better than 0.003 °K/h. Magnetic shielding is provided by two concentric μ-metal shields outside the Dewar plus two concentric 0.002-in. lead-foil cylinders in the helium bath. These latter shields are, of course, superconducting and serve to stabilize the low field in the vicinity of the voltmeter.

The Josephson junctions are prepared by evaporating the desired metal through a mask onto glazed alumina substrates. After oxidation and a second evaporation the crossed-film geometry shown in Fig. 2 results. Film thicknesses are typically 1000 Å.

Contact is made to the samples using pure indium solder. The two upper films of the junctions are connected with a 0.020-in. Pb wire. Any volt-

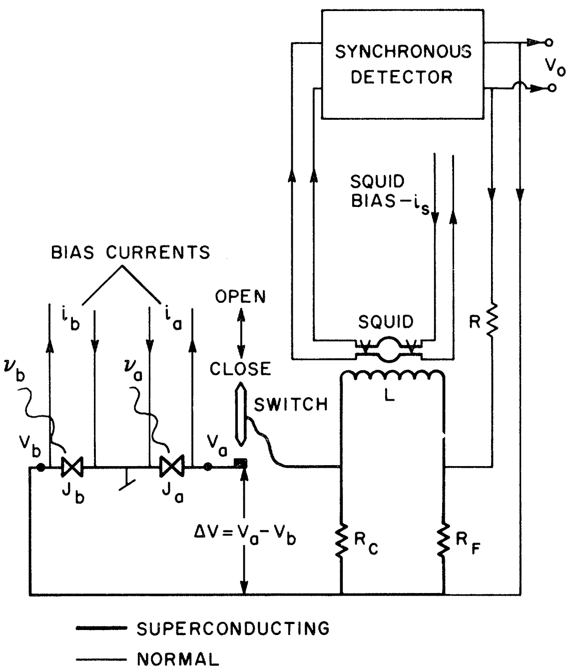

FIG. 1. Circuit to compare constant-voltage steps on two Josephson junctions J_a and J_b which are irradiated with microwaves at frequencies ν_a and ν_b. The SQUID detects any current in coupling coil L and thus serves as a galvanometer in the potentiometer consisting of arms R_F and R_c. A signal ΔV across R_c actuates control voltage V_0, which is fed back through resistive network R to null the potentiometer. The control voltage is recorded and easily calibrated in terms of volts across R_c. The modulation coil has been omitted and the room-temperature amplifiers have been represented as a simple detector.

age difference between the biased junctions then appears as a voltage difference between the lower two films. The superconducting leads to the switch and the voltmeter consist of Pb wire and radio solder. The latter is used because it is readily spot welded to the niobium-foil leads of the voltmeter.

The mechanical switch which connects the junctions to the voltmeter consists of a copper point tinned with soft solder that is pushed into a flattened piece of soft solder. The point is operated from the top of the Dewar by means of a stainless-steel tube with a brass extension. The point is insulated from this push rod and from the guide tube by a delrin bushing. A piece of niobium foil is spot welded to the tinned copper point. This foil is then spot welded to one of the niobium-wire leads of the voltmeter. The foil is thin and remains flexible at liquid-helium temperatures, so the switch can be operated without breaking the leads. Contact to the stationary portion of the switch is made by soldering one of the Pb wires from the sample to the solder. The resistance of the closed switch and leads measured with the junctions in a dc state is less than 10^{-11} Ω and the voltage measured by the voltmeter is entirely due to any signal at the samples. However, as discussed below, the mechanical nature of the switch can introduce a systematic error in the voltage zero.

The two samples are mounted in separate waveguides (Fig. 2). The junction dimensions, 0.5×0.8 mm, are chosen so that they are resonant at 10 GHz. The frequency of the incident radiation can be varied to maximize coupling. The common side of the two waveguides is butted against the shorting termination. This and judicious use of microwave absorbers around the rest of the end of the guides afford at least 30 dB of isolation between the two samples.

The microwave radiation is provided by two x-band klystrons, which are phase locked with separate Dymec Model No. 2654 frequency standard synchronizers (Fig. 3). A stable reference signal at 5 MHz for the system is provided by a Hewlett Packard Model No. 107BR crystal oscillator with a short-term stability of 1.5 parts in 10^{11}. However, the magnitude of the uncertainty in the difference frequency is determined by the stability of the klystrons with respect to the oscillator. For these klystrons the difference frequency between the two klystrons is stable to about 5×10^{-3} Hz or $\Delta\nu/\nu \sim 5\times10^{-13}$.

To obtain a finite difference frequency $\Delta\nu$ between the two klystrons, an external intermediate frequency at 30 MHz$\pm\ \Delta\nu$ is used for one of the synchronizers. The external i.f. signal is supplied by a General Radio Model No. 1164A frequency synthesizer which is also referenced to the stable oscillator. The difference frequency can easily be varied from 0.01 to 1 MHz either manually or using the voltage-controlled-oscillator (VCO) capability of the synthesizer.

To measure voltage differences of less than 10^{-12} V between corresponding steps on two junctions, a niobium-foil SQUID is used as a null detector in the potentiometer circuit shown in Fig. 1. In this case the well-known field-dependent voltage periodicity of the SQUID is used to measure the current produced in the coupling circuit L by a voltage across R_c. The magnetic field on the SQUID is modulated by a separate coil for the purpose of synchronous detection. The dc output voltage from the synchronous detector (Princeton Applied Research Model No. JB-4 lock-in amplifier) is then fed back across R_F with the appropriate gain to close the loop and null the potentiometer. The closed-loop output V_0 from the lock-in amplifier is then proportional to the voltage across R_c. The resistances in the potentiometer arms are equal, $R_F \approx R_c \approx 8\times10^{-5}$ Ω. They are made of 0.025-

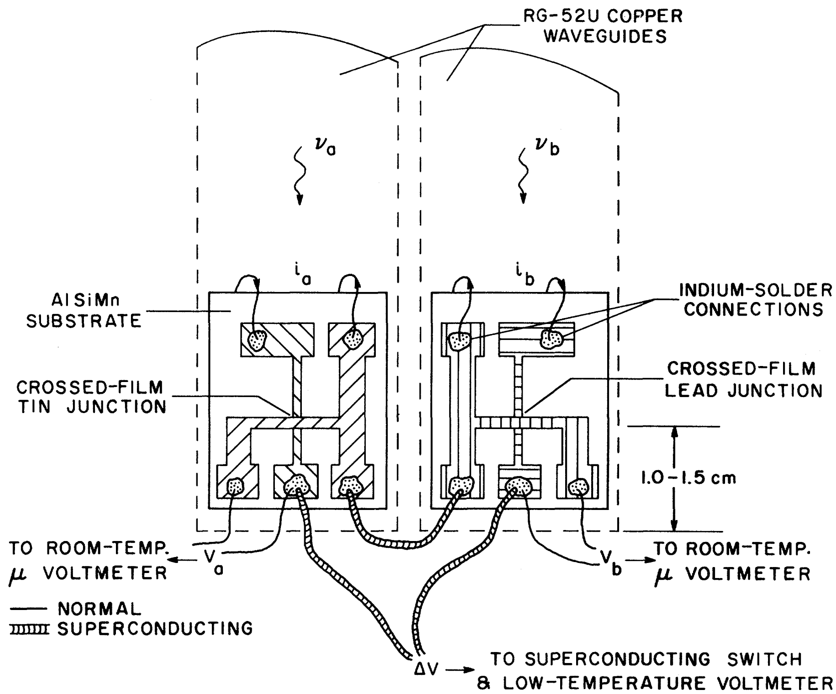

FIG. 2. Sample configuration and alignment in waveguides.

in. brass shim stock tinned with solder at each end. With visual averaging of the recorder output, the sensitivity of the voltmeter is typically better than 2×10^{-13} V.

A loop equation for the voltmeter can easily be written down in terms of the transfer functions of the various components and then solved for the output voltage V_0.[7] In this case we are ultimately interested only in the dc response of the system, so the transfer functions reduce to constants. In the limit of high gain and long time, the closed-loop control voltage takes the general form

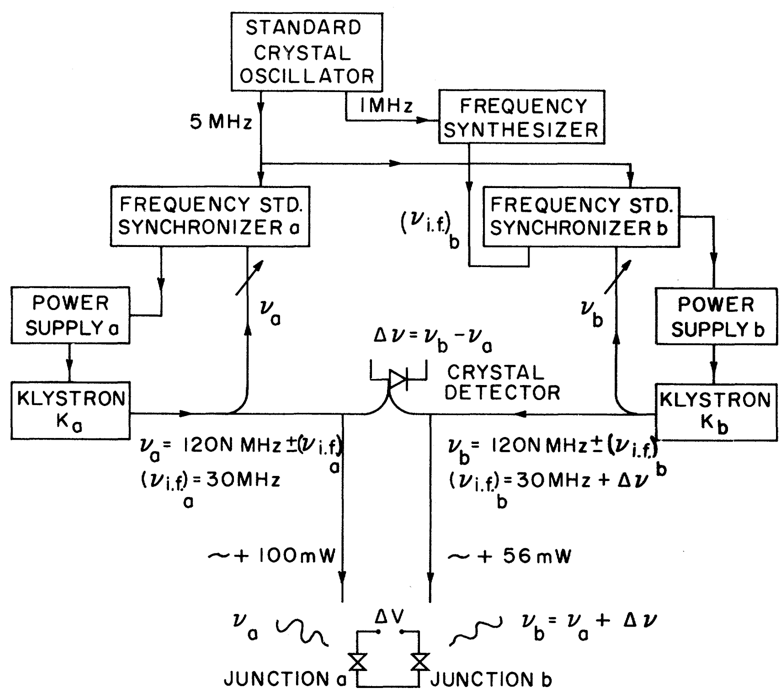

FIG. 3. Microwave-stabilization scheme. The synchronizers stabilize the klystrons with repect to the crystal oscillator at $\nu = 120N$ MHz $\pm\ \nu_{i.f.}$, where N is the chosen harmonic of 120 MHz and $\nu_{i.f.}$ is the intermediate frequency in the phase-lock system. The synchronizer for klystron K_a uses its 30-MHz internal intermediate frequency. The intermediate frequency for the other klystron $(\nu_{i.f.})_b = 30$ MHz $+\Delta\nu$ is provided by the frequency synthesizer. The two klystrons are then phase locked at frequencies $\Delta\nu$ apart.

$$V_0 = (1/\beta_1)(G\Phi + \beta_2 v), \quad (1)$$

where Φ is the ambient magnetic flux through the SQUID, v is the voltage signal across R_c, β_1 represents the attenuation of the feedback network, G is a magnetic-flux-to-current conversion factor, and β_2 is the conduction of the coupling loop. Thermal voltage terms are small compared with the minimum detectable signal and hence have been ignored in this simplified analysis.

With the switch open and $R_c = R_F$, the current through the feedback loop splits equally between the coupling loop and the feedback resistor. However, when the superconducting switch is closed the calibration resistor is shorted out and all the current goes through the coupling coil. Thus the gain of the system changes discontinuously when the switch is closed and we have $\beta_1' = 2\beta_1$, where the primed and unprimed variables now denote the closed- and open-switch positions, respectively. The conversion factors G and β_2 do not change when the switch changes position. However, if the magnetic field at the SQUID is dependent on the physical location of the switch, then the two positions would yield different ambient-field configurations. Thus we have $\Phi' = \Phi - \delta\Phi$, where $\delta\Phi$ is the difference in flux through the SQUID for the two switch locations.

The output voltages for the two switch positions are then

$$V_0 = (1/\beta_1)(G\Phi) \quad (2)$$

and

$$V_0' = (1/2\beta_1)[G(\Phi - \delta\Phi) + \beta_2 \Delta V], \quad (3)$$

where the open-switch signal v is zero, the closed-switch signal v' is ΔV, and an ambient flux Φ is assumed. If the residual field remains constant during the switching time the voltage difference between the junctions is given by

$$\Delta V = (\beta_1/\beta_2)(2V_0' - V_0) + \Delta,$$

where $\Delta = G\delta\Phi/\beta_2$ is the signal due to any change in field at the SQUID associated with switching. Thus, the difference of two measurable absolute quantities $2V_0'$ and V_0 yields a value for any voltage from the junctions plus a possible constant offset. The signal is easily calibrated. The value for β_2 is found by measuring R_F. Then, given this resistance and R_c, β_1 is determined by measuring the change in V_0 for a known current through R_c.

The ambient flux Φ can be nulled by an appropriate dc current in the modulation coil. Then zero output voltage corresponds to zero signal voltage for one position of the switch. If the field is not nulled, the small signal $(\beta_2/\beta_1)\Delta V$ becomes the difference between two relatively large signals $2V_0'$ and V_0. Furthermore, without the field-zeroing procedure, the transients associated with the change in gain when switching are so large that they can cause the SQUID to jump from one stable lock point to another. As long as the net ambient flux through the SQUID is small, $\Phi \leq \frac{1}{10} hc/2e = \frac{1}{10}\Phi_0$, the voltmeter remains locked when switched, and the output voltage is not large.

Although there are long periods of stability in the output voltage, generally there is a slight drift in one direction associated with the changing helium level. The magnitude of this drift is about 5×10^{-13} V/min, which corresponds to a field change at the SQUID of about 10^{-7} G/min. The actual duration of each run between helium transfers is about 2 h. After the period of relative stability characterized by the small unidirectional drift, a substantial increase in drift rate occurs, indicating that the superconducting shields have started to go normal.

EXPERIMENTAL RESULTS

The sensitivity of the voltmeter and the stability of the voltmeter during switching make a voltage comparison between the constant-voltage steps possible. The resistance of the steps is measured by varying the bias current of one junction and looking for a voltage change relative to the other junction. The upper limit for the resistance of the $n = 15$ step on a shorted tin junction was $R_{Sn} \leq 7\times 10^{-11}$ Ω and for a lead junction $R_{Pb} \leq 10^{-8}$ Ω. In each case no voltage change was detected over the width of step and the limit is set by the maximum change in bias current. The steps are typically tens of microamps wide and the small current ($\leq 10^{-8}$ A) drawn through the voltmeter is not sufficient to unbias the junction. Thus the induced steps are wide enough and flat enough to make a voltage comparison at the 10^{-13}-V level meaningful.

The frequency-dependent voltage difference between two tin junctions biased on the $n = 5$ step at 9.870 GHz is shown in Fig. 4. The frequency on one of the junctions was varied at a constant rate and the voltage output V_0' from the lock-in amplifier recorded as a function of the frequency difference between the two junctions. The variable frequency was provided by driving the VCO of the intermediate frequency synthesizer with a 1-min triangle function. The frequency difference $\Delta\nu$ was measured as a beat note between the two microwave signals. The rapid drop in output voltage at the end of the triangle sweep is real and due to a spurious cutoff of the function generator signal. The measured slope of the voltage-output–vs–frequency-difference plot is

$$\Delta V/\Delta\nu = 1.06 \pm 0.05 \times 10^{-14} \text{ V/Hz},$$

which is in good agreement with the predicted value of 1.034×10^{-14} V/Hz. The 5% experimental error is primarily due to difficulties in calibrating the voltmeter. Measurements of the voltage change when the frequency is changed by 200 or 1000 Hz also give values for $\Delta V/\Delta\nu$ that are within a few percent of the accepted value of $nh/2e$. If the frequency on the opposite junction is varied, the voltage reverses.

From these observations, it is clear that the voltmeter can observe a frequency-dependent voltage difference between two corresponding constant-voltage steps. The sensitivity of the voltmeter in Fig. 4 in frequency units is about $\Delta\nu = \pm 4$ Hz or about 4 parts in 10^{10}. However, this high resolution is with the switch closed and represents only a relative voltage measurement.

In order to detect any inherent difference between the steps on two junctions, the true voltage null must be established. This can be done by comparing the open- and closed-switch voltages. The voltage signal from the junctions ΔV is then given by Eq. (4) and the voltage null is determined by the recorder zero. Thus, the procedure for taking data to establish the absolute difference is as follows. With the two junctions biased on the same step, the switch is opened and closed and the absolute open (V_0) and closed (V'_0) output voltage recorded. The signal is then computed as $V_s = 2V'_0 - V_0$. The calibration of V_s is easily accomplished either in terms of frequency or volts at the SQUID. The resultant values for v are then averaged over the duration of the run of about 15–20 operations of the switch. The quantity $(\beta_1/\beta_2) \times (2V'_0 - V_0) = \Delta V + \Delta$ has been plotted in Fig. 5 for two runs comparing the $n = 15$ steps of a lead junction and a shorted tin junction biased at 9.870 GHz. Several runs comparing the voltage between the unbiased junctions are also included. The unbiased points were taken with the microwaves and currents turned off. All the points in Fig. 5 represent data from one set of junctions using a simple on-off switch. The error bars represent one standard deviation from the mean value for m operations of the switch. During run 305-306, the switch was opened and closed four times with a 10-Hz frequency difference on the two junctions. The observed change in output for this signal voltage agrees well with the expected change from the zero-frequency difference signal.

The resolution for any one run in Fig. 5 is about ± 10 Hz or 1 part in 10^9. However, for all the runs, whether the junctions are biased or not, there is an offset Δ. In most runs this offset is about 15 Hz or 5×10^{-13} V, but in one run (419a) Δ is about 15×10^{-13} V. The zero-voltage offset changed dramatically downward during run 419, and hence the data can be split into two distinct points. The change took place in an interval between two switchings and occurred while the ambient field was rezeroed with the switch closed. This procedure results in a large transient current through the switch.

Several other runs were attempted with different junctions and a three-position switch. Although adequate steps could not be induced on the current-voltage characteristics, voltage comparisons of unbiased junctions and of a superconducting short could be accomplished. In all these cases there is a zero-voltage offset of the same sign and magnitude as the previous result. This indicates the existence of a systematic voltage error associated with the apparatus.

The systematic voltage offset is relatively independent of the microwave-power level, junction bias current, SQUID bias current, and different switches. The drift and uncertainty in the room-temperature electronics are too small to account for the observed offset. The measured uncertainty in the recorder null is well below the noise level of the output signal and corresponds to an uncertainty in the zero at the SQUID of less than 3×10^{-14} V.

The voltage offset is a real signal originating at

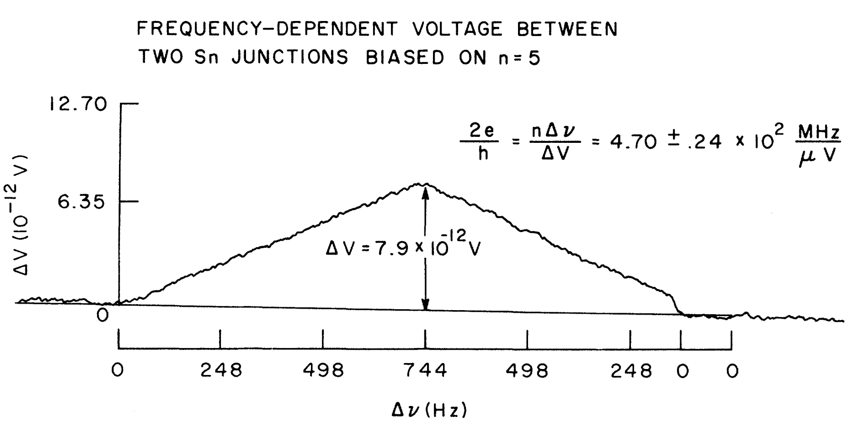

FIG. 4. Frequency-dependent voltage difference between two tin junctions biased on the $n = 5$ step at 9.870 GHz. The rapid drop in voltage at the end of the triangle sweep is due to a spurious cutoff of the voltage controlling the difference frequency.

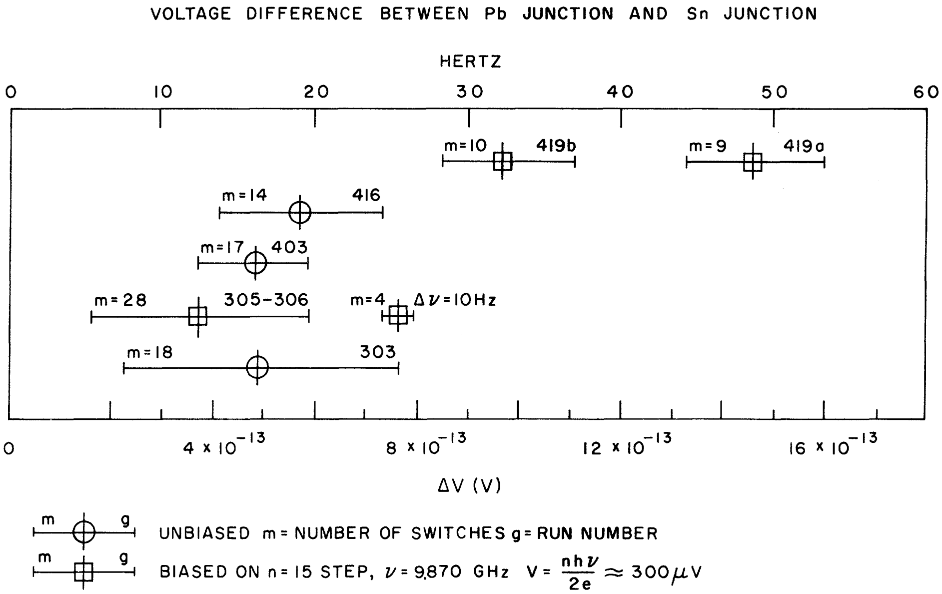

FIG. 5. Measured voltage difference between the $n=15$ steps of a Pb junction and a weakly shorted Sn junction irradiated with microwaves at 9.870 GHz. The measured voltage between the unbiased samples is also shown. The offset from 0 V is discussed in the text.

the SQUID. Inspection of the apparatus at room temperature indicates considerable displacement (1–3 mm) of the switch and leads between the two switching positions. Several things point to this displacement as the source of the offset. In some runs with the sophisticated switch the amount of offset is correlated with the amount of pressure on the switch just before a change in position. Inspection shows that the amount of displacement is directly related to the pressure. In addition, a change in output voltage was observed when the switch was moved without making contact. This indicates a signal that is dependent on the location of the switch. We conclude that this signal is the source of the systematic offset and furthermore that the spurious signal is magnetic in origin. The macroscopic inhomogeneity of the leads indicates the possibility of trapped flux. Direct evidence that the spurious field is due to trapped flux is the aforementioned abrupt change in offset observed during run 419. In this case, the large transient associated with rezeroing was conceivably large enough to force some of the trapped flux from its pinning sites.

A magnetic field change of 4×10^{-7} G ($\sim 0.08\Phi_0$) at the SQUID is necessary to account for the observed offset. Rough estimates of the magnitude of the trapped field necessary to produce this change seem exceedingly large. Nevertheless, trapped flux in the leads is still the most plausible source for the voltage offset. A field due to a magnetic impurity is unlikely because of the abrupt change in offset after a large current was passed through the leads. A change in microwave pickup at the SQUID due to movement of the leads is inconsistent with the observed constancy of the zero offset whether microwave radiation is present or not. Because of the relatively large error bars on the data points, the presence of these two effects cannot be discounted. However, if present, they are small in magnitude compared to the total offset and cannot totally explain the observed behavior. Experiments with a better shielded switch are planned to confirm and, if possible, eliminate the source of the voltage offset. If these are successful, then the addition of a more sensitive voltmeter and the use of higher-order steps could increase the relative sensitivity of the experiment by an order of magnitude or more.

We conclude from Fig. 5 that, even when the offset is included, the upper limit for a voltage difference between the $n=15$ induced steps in Pb and Sn junctions is 1.6×10^{-12} V. This corresponds to a relative uncertainty of about 5 parts in 10^9. The limited amount of data makes any smaller limit tentative without more experiments under varying conditions and with different materials. However, if the offset is, in fact, considered a systematic error, then the limit can be reduced to less than $\pm 3\times10^{-13}$ V or 1-part-in-10^9 uncertainty for the $n=15$ step.

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Specific-Heat and Magnetic Measurements in Superconducting Ta-Nb Alloys*

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The specific heats of Ta and the Ta-Nb alloys $Ta_{0.963}Nb_{0.037}$, $Ta_{0.86}Nb_{0.14}$, $Ta_{0.595}Nb_{0.405}$, and $Ta_{0.42}Nb_{0.58}$ were measured from 1.5 to 7 K in both normal and superconducting states. The electronic specific-heat coefficient γ and the Debye temperature Θ_D were obtained from normal-state measurements below 4.2 K. The electron-phonon coupling factor λ and the bare-band-structure density of states at the Fermi surface $N_{bs}(0)$, calculated from these results using McMillan's theory, show a smooth increase with increasing Nb concentration. The values of λ are also compared with the predictions of Hopfield's theory. The thermodynamic critical field $H_c(T)$ for these intermediate coupling alloys was determined by integrating the difference between the normal and superconducting specific heat $\Delta C(T)$. A comparison with independent values of $H_c(T)$ obtained from magnetization measurements carried out on the *same* samples shows agreement at low temperatures and suggests the magnetic measurements tend to emphasize sample inhomogeneities particularly near T_c. Finally, λ is compared with other indicators of strong coupling in superconductors such as the deviation of $H_c(T)$ from T^2 behavior, $\Delta C/\gamma T_c$, $2\Delta(0)/k_BT_c$, and $H_c^2(0)/\gamma T_c^2$ determined for these alloys from the specific-heat measurements.

I. INTRODUCTION

The comparison of measurements of the electronic specific-heat coefficient γ to calculations of the bare-band-structure density of states at the Fermi surface $N_{bs}(0)$ is complicated by the ubiquitous and often large electron-phonon enhancement factor λ. However, recent semiempirical calculations by McMillan[1] permit quantitative estimates of λ for superconducting metals and alloys from specific-heat and critical-temperature measurements. McMillan also shows that when λ is estimated in this way, good agreement is found between measured values of γ and those predicted from band-structure calculations by Mattheiss[2] together with the rigid-band model for transition-element alloys within a given row of the Periodic Table.

In this paper we present an experimental study of the effects of alloying transition elements from a given column of the Periodic Table where the rigid-band model is known to be unsatisfactory. In order to compare McMillan's λ with other indicators of coupling strength, careful specific-heat measurements were taken in the superconducting as well as the normal state. Furthermore, the calorimeter was designed to utilize samples which were of suitable size and shape for magnetic measurements, allowing us to compare parameters measured by both methods on the same samples. The Ta-Nb system was chosen as one of the simplest systems available for this purpose. The system forms a body-centered-cubic solid solution with very nearly identical conduction-electron density for all alloy concentrations. We have concentrated on the Ta-rich end of the Ta-Nb system and compared our results with those of a similar study reported by Kimura *et al.*[3] on the Nb-rich alloys. The present specific-heat measurements were of sufficient accuracy to allow an independent determination of the thermodynamic critical field as a function of temperature $H_c(T)$ by numerical integration of the thermal data. Magnetization measurements were subsequently made on some of the same samples and the values of $H_c(T)$ obtained by integration of the magnetization curves were in agreement within experimental error with the values obtained by integration of the specific heat.