metals in some manner unassociated with the superconducting state or absorption by the very complicated amorphous transitional-metal oxides involved. The variety of possible absorption mechanisms and the uncertainty concerning the physical state of the junctions make it useless to speculate further upon the causes of these particular temperature-independent features at this time. It is important to know that they do exist and that they must be reckoned with in any work similar to this.

IV. CONCLUSIONS

This work and that done previously[1,8] indicate that reasonable superconducting tunneling behavior can be obtained with point contacts, and that this experimental method should prove very useful. Some of the experimental complications which arise have been pointed out. We have concluded that the subharmonic structure occurs simultaneously with metallic shorts in the junctions. It appears very probable that superconducting weak-link behavior is necessary for the occurrence of subharmonic structure; however, we were unable to determine if it is sufficient. It could not definitely be determined whether electron tunneling is necessary or simply incidental, although it was clearly present during almost all the time that subharmonic structure existed. No combination of the various physical mechanisms which have been proposed could give an explanation consistent with our observations on both like- and unlike-metal junctions. For a satisfactory explanation, more discriminating experiments must be performed and new ideas must be sought.

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Temperature and Magnetic Field Dependence of the Subharmonic Structure in Pb-Pb Superconducting Tunneling

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Peaks occur in the I-V characteristics of lead–lead-oxide–lead superconducting tunnel junctions at submultiples $2\Delta/n$ of the full energy gap 2Δ. This "subharmonic structure" has been studied in detail for its dependence on temperature and magnetic field. Its position varies with temperature identically as the energy gap does. The widths of the subharmonic peaks are independent of temperature. Magnetic fields are found to reduce the amplitudes and increase the widths of the structure. Evidence is presented that none of the present theories explains the phenomenon satisfactorily.

INTRODUCTION

The use of an electron tunneling technique for the study of the energy gap of superconductors was introduced by Giaever and Megerle[1] in 1961. In this method, a metal-insulator-metal thin-film "sandwich" is formed by evaporation and oxidation of the metal. If one or both of the metals are superconductors, an easily measured structure occurs in the current-voltage characteristic at voltages equal to the sum and the difference of the half-gaps, Δ_1 and Δ_2, of the superconductors. When this tunneling method was used to investigate junctions with lead or tin on both sides,[2,3] an unexpected additional structure was observed. In addition to the rise in current at the full gap 2Δ, small peaks of excess current appeared on the I-V curve at submultiples of this value $2\Delta/n$, where $n=2, 3, 4, \ldots$. These peaks have been called subharmonic structure (SHS).

There have been several suggestions[2–8] of possible mechanisms for the SHS. The most thoroughly investigated possibilities have been those of a connection with metallic shorts through the insulator, or else with phenomena related to the Josephson effect. A study of multiparticle tunneling[9] showed that this mechanism

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gave results of the wrong order of magnitude. No clear, consistent picture has emerged from the combination of the results of the various experimenters.[2–6] No satisfactory explanation exists for SHS. The present project was undertaken with the purpose of further investigating the properties of these tunnel junctions. This work dispels some of the contradictions contained in previous results, which prevent a theoretical understanding of the phenomenon.

EXPERIMENTAL METHODS

Samples were made by evaporating a longitudinal lead strip onto a clean glass substrate (see Fig. 1). The lead strip was 1 mm wide and roughly 800 Å thick. It was deposited in a vacuum of less than 3×10^{-6} Torr. After deposition, the top of the strip was oxidized to form an insulating barrier, and then three lead cross strips were evaporated. Each cross strip was also 1 mm wide and about 800 Å thick. The oxide layer was found by capacitance measurements to have an effective thickness of roughly 15 Å.

The oxidation process depended on three parameters: the duration of exposure to air, the temperature, and the humidity. Oxidizing for $1\frac{1}{2}$–2 h in an oven at 55°C yielded at least one usable junction on every substrate made as long as the relative humidity at 25°C was between 30% and 35%. An attempt to counteract the effect of lower humidity by increasing the temperature to 60°C led to an anomalous junction. This junction did not exhibit normal tunneling behavior and will be discussed below. Few authors have described the details of their oxidation procedures, but Rowell and Kopf[10] mention using a temperature of 50°C in a stream of dry oxygen for 5–30 min.

The samples were mounted at the end of a stainless-steel tube so that they could be placed in liquid helium either inside the storage Dewar or in an experimental Dewar for working at large magnetic fields or temperatures below 4.2°K. Magnetic fields of up to 1000 G could be applied in the storage Dewar with a copper solenoid placed around the sample. A carbon composition resistor (1000 Ω at room temperature) was used as a thermometer.

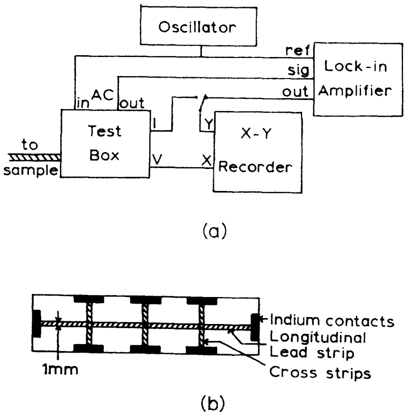

FIG. 1. Experimental arrangement. (a) Block diagram of the measuring circuit; (b) sample configuration.

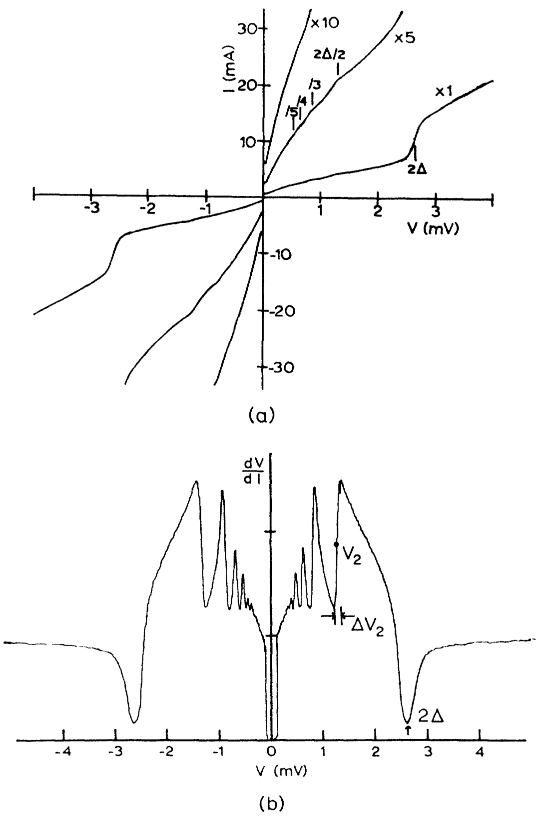

FIG. 2. (a) Typical *I-V* characteristic of Pb-PbO-Pb tunneling junction. The current scale near the center has been expanded to show the subharmonic structure (SHS) at $2\Delta/n$. (b) Derivative (dV/dI versus V) curve for a typical sample. SHS is more clearly visible than in the *I-V* curve itself. The structure is symmetric about $V=0$.

A block diagram of the measuring apparatus is shown in Fig. 1. The test box could select any one of the three junctions or a standard resistor for calibration. A ten-turn Helipot in the test box was used to vary the dc current bias. An oscillator supplied a small modulation signal of several microamperes at a frequency of 5400 Hz to the junction being studied. The oscillator simultaneously supplied a reference signal to the lock-in amplifier (Princeton Applied Research Model 121). Such an arrangement allows one to plot either the current I or the derivative (dV/dI) against the bias voltage V.[11] Batteries were used for both the solenoid and bias supplies in order to avoid problems associated with ripple and ground loops.

One junction on each sample substrate usually showed the structure more clearly than the other junctions and

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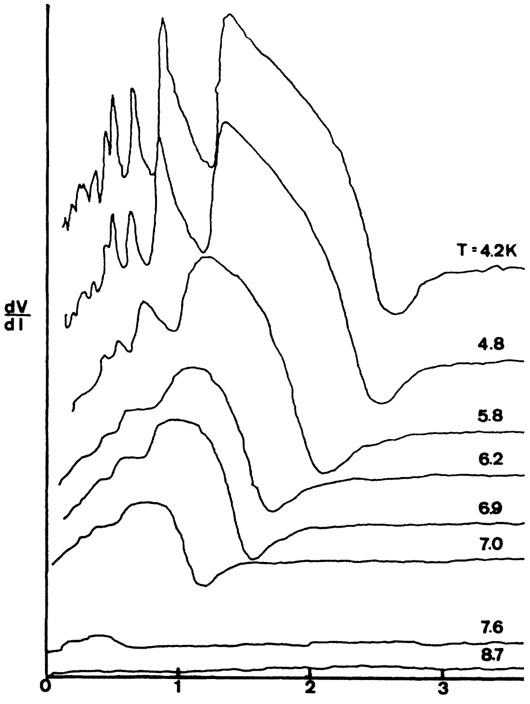

FIG. 3. Derivative curves (dV/dI versus V) for a typical sample for various temperatures. The dependence of the SHS is the same as that of the energy gap 2Δ. No magnetic field is present.

was selected for study. Derivative curves were then taken on this junction, either at constant temperature for various magnetic field strengths or at constant field for various temperatures. Temperatures above 4.2°K were attained by raising the sample into the gas above the liquid helium and, at zero magnetic field, could be maintained fairly constant over a period of several minutes. The presence of the copper solenoid around the sample helped stabilize the temperature when no current was being supplied to the solenoid.

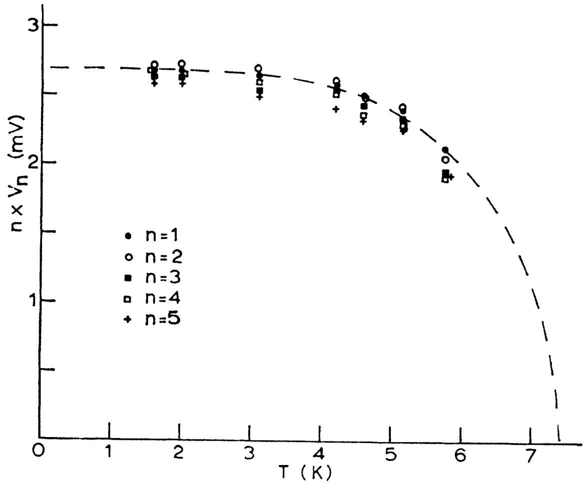

FIG. 4. Temperature dependence of the energy gap 2Δ and of the positions of SHS in voltage V_n for $n=2$–5. Plot shows $n\times V_n$. The accuracy is best for small n. No magnetic field is present.

EXPERIMENTAL RESULTS

Figure 2(a) shows the current-voltage characteristic of a typical junction at 4.2°K. The current scale near the center is magnified to show the subharmonic peaks more clearly. A derivative curve (dV/dI versus V) is shown in Fig. 2(b). The structure at $2\Delta/n$ is defined much more clearly in the derivative. Both the I-V and derivative curves are completely symmetrical. The minimum at 2.64 mV was taken to measure the full energy gap in Pb, 2Δ. The midpoints of the steeper (low-voltage) sides of the peaks were taken to indicate the positions of the structure on the I-V curve. Positions chosen in this way do not agree precisely with the values of $2\Delta/n$, probably because of the sloping background current characteristic. The disagreement is worse for higher n. For example, in Fig. 2(b), the structure at $n=5$ is observed at 0.49 mV instead of at 0.53 mV ($=2\Delta/5$); the structure at $n=2$ occurs at 1.31 mV instead of 1.32 mV. A series of derivative curves (dV/dI versus V) is shown in Fig. 3, for one junction at several temperatures. The temperature dependence of the peak positions in Fig. 3 can be seen from Fig. 4 to

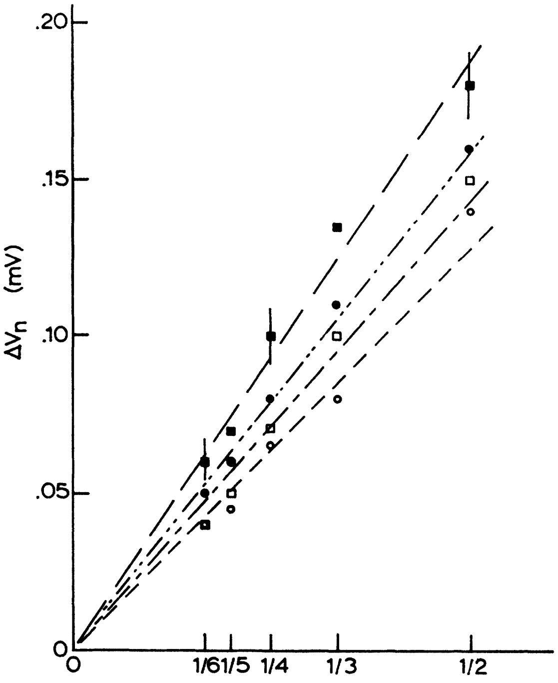

FIG. 5. Widths $\Delta V_n=(V_{\max}-V_{\min})$ for nth peak plotted against $1/n$ for four sample junctions. The temperature is 4.2°K; no magnetic field is present.

be the same as the temperature dependence of the full energy gap, 2Δ ($n=1$).

A V_n may be defined as the central voltage of the nth peak and ΔV_n as the difference in voltage between the maximum and minimum of the derivative at the nth peak (see Fig. 2). This ΔV_n is seen in Fig. 5 to be approximately proportional to $1/n$. Since V_n is also proportional to $1/n$, the ratio $V_n/\Delta V_n$ is the same for all peaks for a given sample at given temperature and magnetic field. The widths of ΔV_n of the peaks are, within the experimental accuracy, independent of the temperature. The peaks are less clearly defined at higher temperatures because the gap grows smaller and the thermal background current becomes larger.

The effect of a magnetic field is shown by the series of derivative curves in Fig. 6. The magnetic field lies in the plane of the junction. The adjustment of this angle is not critical. Several observations may be made. First the SHS is obliterated by sufficiently large magnetic fields (about 1000 G for this particular sample). Second, the energy gap is still reasonably well defined at this field strength and requires about 1600 G to be completely removed. Third, the SHS is sharper for $B=225$ G than for $B=0$. This phenomenon was not observed in all samples. A final observation, not illustrated in Fig. 6, is that flux trapping occurred in this sample, equivalent to an external field of about 400–500 G. Some junctions did not exhibit any flux trapping at all, however.

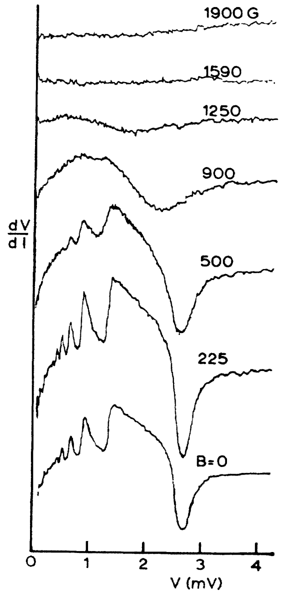

FIG. 6. Derivative curves (dV/dI versus V) for a typical sample for various magnetic field strengths. The field strength in gauss is indicated for each curve. The SHS disappears at about 1000 G; the energy gap disappears at about 1600 G. The temperature is 4.2°K.

Figure 7 shows the amplitude of the derivative, defined as the height of the first derivative (dV/dI) from minimum shoulder to maximum shoulder for $n=2$, plotted against field strength. This curve gives an idea of the rate at which the structure disappears with increasing field strength. It also allows an extrapolation to the field required to obliterate the structure completely.

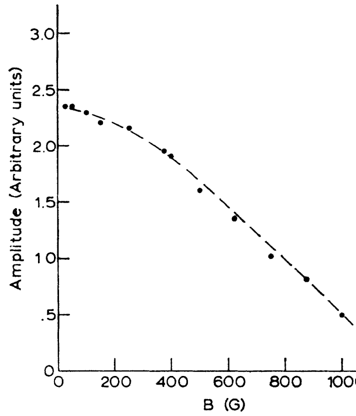

FIG. 7. Amplitude of derivative peak for $n=2$ plotted against magnetic field strength. The curve shows the rate at which the SHS goes away, and allows an extrapolation to the field strength at which the SHS disappears completely. The temperature is 4.2°K.

The dependences of V_n and ΔV_n upon magnetic field are shown in Fig. 8. This result disagrees with previous reports that the locations and widths of the peaks are insensitive to the magnetic field. Here we see that the widths are increased by a factor of 2, while the location is shifted only slightly, following the change in the energy gap. These changes are purely magnetic effects and cannot be attributed to any thermal effect.

DISCUSSION

Certain of the results described above have previously been reported by various other authors, in particular Marcus,[2] Rochlin,[5] and Rowell and Feldmann.[6] Some

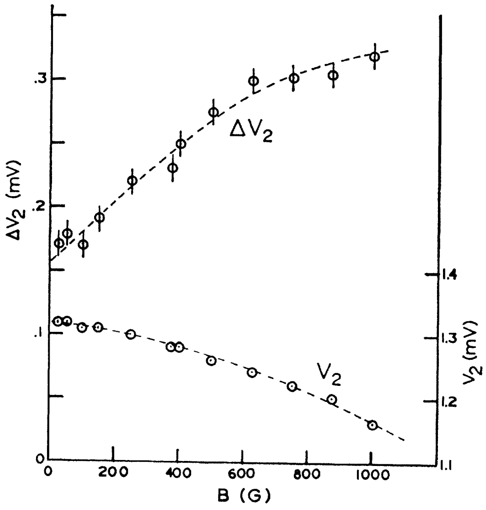

FIG. 8. V_n and ΔV_n plotted for $n=2$ as functions of magnetic field strength. Although the detailed dependences upon magnetic field differed among samples, there was always a sizable increase in the width ΔV_n.

of the present results, however, have not been reported previously. They allow a reinterpretation of previous results, this reconciling some of the contradictory conclusions, and allowing a more unified description of the behavior of the SHS.

Certain previous investigators have given several reasons for believing that the peaks are caused by a tunneling process taking place in regions of the junction where there are metallic filamentary shorts between the metal films.[2,6] Several of the junctions exhibiting SHS in this study showed no measurable zero-voltage current, however, which indicates that no superconducting lead shorts were present at all. Furthermore, several other junctions were observed to have large zero-voltage currents but only very weak subharmonic peaks. These junctions very probably had superconducting shorts, but the shorts only obscured the peaks and did not enhance them. Some investigators have noted that flux trapping has always occurred in their samples showing SHS. Flux trapping might be evidence of imperfections in the oxide layer. While some of our junctions having SHS showed flux trapping, others did not; so the presence of flux trapping appears not to be related to the peaks.

The suggestion has been made[5] that the SHS is caused by a single-particle tunneling effect enhanced at the subharmonic voltages by a mechanism involving the Josephson effect. As evidence for this explanation, the fact is cited that many junctions showed Josephson effects and seemed to be free of any shorts, bridges, or other imperfections. One of our junctions showed subharmonic peaks with no evidence of single-particle tunneling at all. Rowell and Feldmann[6] have also observed a junction in which SHS was present without a single-particle tunneling current. This evidence might suggest that a single-particle superconducting tunneling mechanism may be inadequate as an explanation for the SHS.

Both Rowell and Feldmann[6] and Marcus[2] report that the structure is relatively insensitive to magnetic field. Significant effects were found in the samples studied in this work. The SHS showed the following field dependences:

(1) Both the positions and the widths of the peaks were found to be field-dependent.

(2) The higher-order peaks were noticeably weakened by fields too small to have any effect on the lower-order peaks.

(3) A small magnetic field (about 200 G) sometimes enhanced the magnitude of the SHS.

Other interesting phenomena were found in magnetic field studies. Small "spikes" were sometimes observed in the derivative curves at very low voltages in the presence of magnetic fields of less than 100 G. The field dependence of these spikes was complicated.

Rowell and Feldmann[6] have suggested that because of the lack of observed field dependence of the phenomenon, the SHS could not be due to resonant interactions between the junction cavity and the ac Josephson field. The observations noted above reduce the level of assurance with which this statement can be made, since the observations suggest that the mechanism involved in the SHS is affected by magnetic fields.

CONCLUSION

The following conclusions have been obtained from this study of subharmonic structure:

(1) The structure is present at submultiples of the full energy gap ($2\Delta/n$) in lead-lead-oxide-lead tunnel junctions.

(2) The position of the SHS has a temperature dependence identical to that of the energy gap, throughout the temperature range below T_c, the critical temperature of lead. Previous studies have apparently been limited to temperatures below 4.2°K.

(3) The widths of the peaks are proportional to $1/n$, as reported previously.[2]

(4) The widths of the peaks are independent of the temperature below T_c. This result extends that of previous work done below 4.2°K.

(5) The SHS is destroyed by magnetic fields smaller than those required to destroy superconductivity in the lead, as reported by Marcus.[2]

(6) The SHS can occur in junctions with or without flux trapping or superconducting shorts.

(7) The widths of the peaks increase with increasing magnetic field. This contrasts with the temperature independence of the widths.

(8) The structure sometimes is enhanced by a small magnetic field.

(9) Spikes appear in the derivative (dV/dI versus V) curves at low voltages in the presence of small magnetic fields. Their amplitude depends on the field strength in a way that suggests that a resonance phenomenon could be involved.

No single explanation offered so far is consistent with all of these conclusions. The failure of previous investigators to agree on the properties of tunneling junctions showing SHS may have been due to differences in emphasis rather than disagreement in experimental results. The present paper has been an attempt to produce a unified picture of the SHS and its temperature and magnetic field dependence. It is hoped that further theoretical consideration of the phenomenon can now be carried on productively.