Excess Currents in Superconducting Tunnel Junctions

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(Received 4 March 1968)

We have studied the excess currents that flow for $V<\Delta_1+\Delta_2$ in two superconductor (S_1–I–S_2) tunnel junctions. The features of interest are "multiparticle" tunneling, which is observed as a current increase at Δ_1 and Δ_2, and the subharmonic effects which occur at $2\Delta/n$ when $\Delta_1=\Delta_2$. Strong evidence is presented that subharmonic junctions contain metallic shorts, and we show that the $2\Delta/n$ series can be observed even in the absence of single-particle tunneling. By extending the study to asymmetrical junctions ($\Delta_1\neq\Delta_2$) we conclude that at present there is no satisfactory explanation of the subharmonic effects.

I. INTRODUCTION

If a tunnel junction, comprised of two superconducting films S_1 and S_2 separated by an insulating layer I, is cooled far below the transition temperature of the films, then the current flowing for applied voltages much less than $\Delta_1+\Delta_2$ (Δ_k being the energy gap of S_k) should be very small compared to that flowing for $V>\Delta_1+\Delta_2$. For example, in Pb at 1°K the number of quasiparticles excited across the gap Δ (1.39 meV) is $\sim N_0 e^{-\Delta/kT}\sim 10^{-7}N_0$. The tunnel current flowing in a Pb–I–Pb junction at 1°K for very small applied biases should be due only to these excited quasiparticles. Thus in a perfect Pb–I–Pb junction at 1°K, the current (I_{SS}) flowing for $V\ll 2\Delta$ should be $\sim 10^{-7}$ of that flowing at the same voltage when the films are normal (I_{NN}). In practice, currents much larger than this are observed, typically 10^{-5} to $\frac{1}{2}I_{NN}$. All such currents greater than the thermally excited quasiparticle currents can be classed as excess currents.

The first measurements of excess currents for $V<2\Delta$ were reported by Taylor and Burstein[1] and, independently, by Adkins.[2] They observed that in certain Sn–I–Sn and Pb–I–Pb junctions the current increased rather abruptly at $\Delta_{\rm Sn}$ or $\Delta_{\rm Pb}$, the size of this increase being $\sim 10^{-4}$ of that at $2\Delta_{\rm Sn}$ or $2\Delta_{\rm Pb}$. In S_1–I–S_2, Taylor and Burstein[1] observed the increases at Δ_1 and Δ_2 in addition to the thermal quasiparticle peak at $\Delta_1-\Delta_2$.

The simplest explanation of this current and the Δ structure was considered in both the above papers, that is, the possibility of normal metal regions existing in the films. Adkins showed that this could not explain the observed currents, although recently Donaldson[3] has shown that even the earth's field trapped in Al–I–Ag junctions can give rise to an appreciable current which can be called the first type of excess current. An alternative explanation was proposed by Schrieffer and Wilkins,[4] who calculated the current due to the destruction of a pair in S_1 and transfer of the two quasiparticles to S_2. This "multiparticle tunneling" accounted qualitatively for the shape of the Δ step, but in order to explain quantitatively the magnitude of the step a "patchy oxide" had to be postulated.

A third type of I–V characteristic for $V<2\Delta$ has been confused with "multiparticle tunneling" for a number of years but it will be discussed separately as it seems in all respects to be a different type of behavior requiring a new explanation. Taylor and Burstein observed that in some of their junctions structure was observed at $2\Delta/3$ and $2\Delta/4$ and suggested that this arose from higher-order multiparticle processes. Using the second-derivative technique, the author[5] observed such structure to at least $2\Delta/12$, and it seemed very unlikely that multiparticle processes would have an observable amplitude to such a high order. Further measurements were made by Yanson *et al.*[6] and by Marcus,[7] who first suggested that two distinct processes were occurring, the Δ structure due to multiparticle tunneling and the $2\Delta/n$ series of structures which have come to be called subharmonic tunneling. This subharmonic tunneling was shown by Rochlin[8] to exhibit considerable fine structure which was interpreted as a number of $2\Delta/n$ series generated by different critical points in an anisotropic gap. In view of the lack of other evidence[9] for anisotropy of the gap in Pb, this interpretation of the data seems open to some discussion.[10]

In this paper multiparticle and subharmonic tunneling will be discussed and the reasonableness of the

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[1] B. N. Taylor and E. Burstein, Phys. Rev. Letters **10**, 14 (1963).

[2] C. J. Adkins, Phil. Mag. **8**, 1051 (1063); Rev. Mod. Phys. **36**, 211 (1964).

[3] G. B. Donaldson, in Proceedings of the Conference on Tunneling, Risø, 1967 (unpublished); Solid State Commun. **5** (1967).

[4] J. R. Schrieffer and J. W. Wilkins, Phys. Rev. Letters **10**, 17 (1963).

[5] J. M. Rowell, Rev. Mod. Phys. **36**, 215 (1964).

[6] I. K. Yanson, V. M. Svistunov, and I. M. Dmitrenko, Zh. Eksperim. i Teor. Fiz. **48**, 976 (1965); **47**, 2091 (1964) [English transls.: Soviet Phys.—JETP **23**, 650 (1965); **20**, 1404 (1965)].

[7] S. M. Marcus, Phys. Letters **19**, 623 (1966); **20**, 236 (1966).

[8] G. I. Rochlin, Phys. Rev. **153**, 513 (1967).

[9] Using their point-contact tunneling technique, H. J. Levinstein and J. E. Kunzler have investigated many orientations of single-crystal Pb and found no anisotropy greater than ±0.03 mV.

[10] A. Zawadowski, Phys. Letters **23**, 225 (1966); M. Ivanchenko, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktsiyu **4**, 358 (1966) [English transl.: Soviet Phys.—JETP Letters **4**, 242 (1966)].

explanations proposed to date considered. Strong evidence that junctions which exhibit subharmonic behavior contain metallic shorts will be presented. New data on junctions which are asymmetric (S_1–I–S_2) will be shown to contradict all the current models used to explain the subharmonic current.

II. EFFECT OF MAGNETIC FIELD AND TRAPPED FLUX

As mentioned above, trapped flux has been identified as the cause of excess currents only in the case of the Al–I–Ag junctions studied by Donaldson.[3] This is presumably due to the low critical field H_c of Al compared to that of Sn and Pb used by other investigators, so that even the earth's field H_E is appreciable compared to H_c. One would expect the fraction of normal material in the film to be $\sim H_E/H_c$ and thus the excess currents $\sim H_E/H_c$. The S–I–M junctions studied by Donaldson are, however, rather uninteresting compared to S–I–S junctions. As discussed by Adkins,[2] the excess current flowing in S–I–S junctions for $0<V<\Delta$ is due to electrons from a normal region in one film passing through the insulator into a normal region in the other film. As the strongly preferred tunneling direction is perpendicular to the films, this current for $0<V<\Delta$ in fact measures the overlap of normal regions on the two sides of the barrier. This current I'_{NN} also flows for $V>\Delta$ but, in addition, we have, for $\Delta<V<2\Delta$, the current I'_{NS} from normal material on one side of the barrier to superconducting material on the other. By measuring I'_{NN} and I'_{NS} as a function of applied field (transverse to the films) one can in fact determine whether the flux penetrates both films and oxide with perfect overlap of the normal regions ($I'_{NN}\propto H/H_c$, $I'_{NS}=0$), whether the fluxoids overlap at random [$I'_{NS}\propto H/H_c$, $I'_{NN}\propto (H/H_c)^2$ for small fields], or whether they avoid any overlap [$I'_{NS}\propto H/H_c$, $I'_{NN}\propto (H/H_c)^n$, where $n>2$]. The measurements of Adkins indicated that little overlap of the normal region occurs, and our own investigation shows, in fact, that overlap seems to be avoided. In view of the work of Giaever[11] on magnetically coupled films, a detailed study of this point for films of different thicknesses and areas seems to be in order.

Gaplessness in the superconducting films, which is also observed as an excess current for $V<2\Delta$, can be produced by parallel magnetic fields,[12] magnetic impurities in the film,[13] an adjacent film of magnetic material,[14] or magnetic impurities in the oxide.[15] Apparent "gaplessness" is observed where the surface of the superconductor is so contaminated that it is superconducting only because of its proximity to the bulk clean material. This is common with junctions made on materials whose superconducting properties are seriously affected by adsorbed gases or those having short coherence lengths.[16] A high density of states at low energies is also present in the normal-metal side of NS proximity layers, although this system is only strictly "gapless" for an infinitely thick N layer.[17]

III. MULTIPARTICLE TUNNELING

The most obvious feature of "multiparticle tunneling" is the rather sudden onset of current flow near Δ. The I-V characteristic for a Pb–I–Pb junction is shown in Fig. 1; and the following features, which are typical of this type of junction showing the Δ structure, are apparent from the plot:

(1) The current at 2.9 mV ($V>2\Delta$) is $\sim 10^3$ times greater than that at 2.4 mV ($V<2\Delta$).

(2) There is rapid increase in current near Δ, with Δ falling roughly in the middle of the current rise. The current for $V>\Delta$ (1.6 mV) is approximately 10 times that for $V<\Delta$ (1.0 meV).

(3) A weak structure occurs at $V<\Delta$ but imagination is required to associate it with the voltage $2\Delta/3$ or $2\Delta/4$. In fact, the increase in current occurs rather smoothly between these two voltages.

Turning now to an examination of the rise in current near Δ, we can see that in fact the point where the current starts to increase rather suddenly is slightly less than 1.2 mV, and the shoulder where the conductance decreases again is $\sim$1.5 mV. If we associate the width of this rise near Δ with smearing of the gap, then we would expect the rise at 2Δ to be spread from 2.4 to 3.0 mV, which is obviously not true. This problem of the width of the Δ rise has not been mentioned

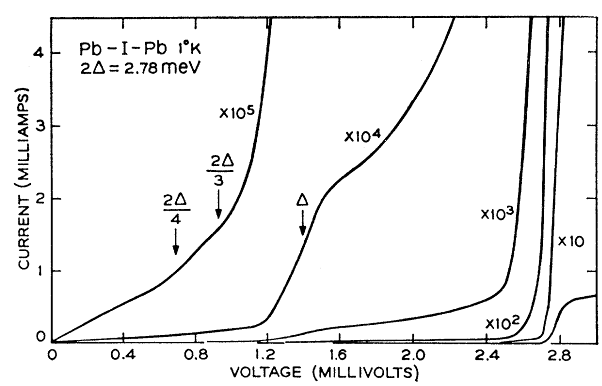

FIG. 1. I-V characteristic of a Pb–I–Pb junction at 1°K. The current scale has been expanded as indicated. We consider this behavior typical of multiparticle tunneling.

[11] I. Giaever, Phys. Rev. Letters **15**, 825 (1965).
[12] J. L. Levine, Phys. Rev. **155**, 373 (1967); J. Millstein and M. Tinkham, Phys. Rev. **158**, 325, (1967).
[13] M. A. Woolf and F. Reif, Phys. Rev. **137**, A557 (1965).
[14] See Ref. 13. Also J. J. Hauser, Physics **2**, 247 (1966).
[15] L. Y. L. Shen and J. M. Rowell, Phys. Rev. **165**, 566 (1968).
[16] For example, tantalum or niobium. See P. Townsend and J. Sutton, Phys. Rev. **128**, 591 (1962).
[17] W. L. McMillan (to be published).

in previous publications and it would be useful to know whether our results are consistent with other observations. In Fig. 2 we have tried to make the point more obvious by plotting parts of the *I-V* characteristic near Δ and 2Δ. One can see that, even in terms of absolute voltage spread, the rise at Δ is broader than that at 2Δ. It could be argued that tunneling from normal metal regions (produced by a magnetic field) into superconducting Pb would give a broad current rise near Δ, but the sharp current onset near 1.2 mV is not consistent with such an explanation. Comparison with *M–I–*Pb junctions reveals that the shape of the rise in current near Δ in such junctions is very different from the shape of the *I-V* characteristic near Δ in Figs. 1 and 2.

An interesting plot which reveals more clearly the relative magnitudes and widths of the effects at Δ and 2Δ is obtained by plotting log*I* versus *V* from 0 to 3 mV (Fig. 3). The increase in current at 2Δ is ×700, that at Δ ×6, and between 2Δ/3 and 2Δ/4 only ×1.2. It seems clear from this plot that, however one estimates the widths of the current increases at Δ and 2Δ, the increase at Δ is appreciably broader (by about 50%) than that at 2Δ. This appears to eliminate the possibility that thermal smearing, or the contribution of thermal phonons to the tunnel current, is responsible for these widths.

The explanation of this tunneling process at Δ was proposed by Schrieffer and Wilkins.[4] The tunneling diagrams of Fig. 4 illustrate (A) the single-particle process where a pair in superconductor 1 breaks up into a quasiparticle in 1 and another which has tunneled into 2, and (B) the "two-particle" process of

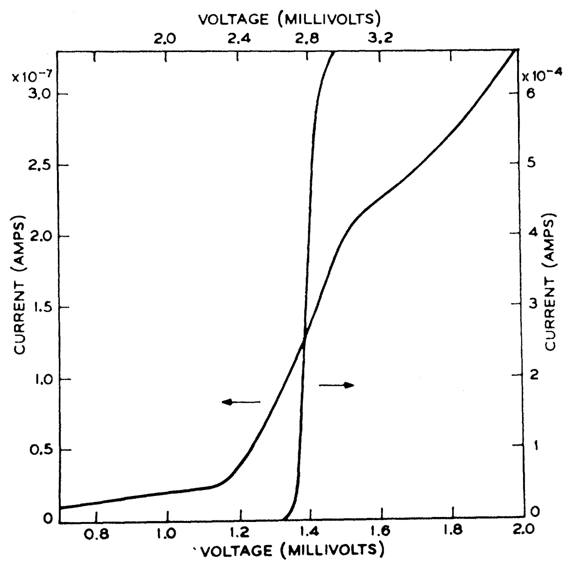

FIG. 2. The parts of the *I-V* plot near Δ and 2Δ for the junction of Fig. 1. The Δ region uses the left and bottom scales, the 2Δ region the top and right scales.

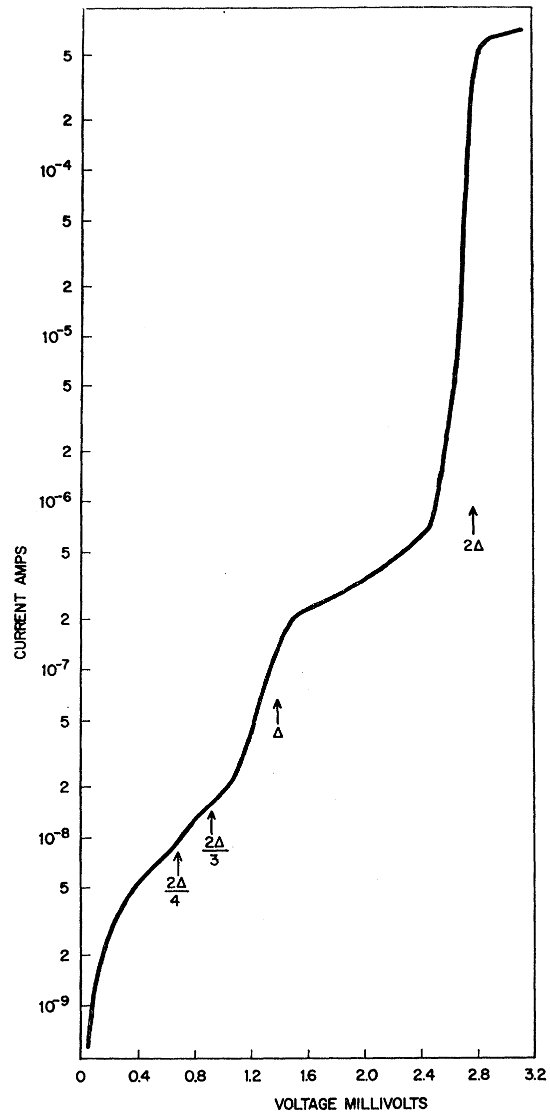

FIG. 3. The *I-V* characteristic of the Pb–*I*–Pb junction of Fig. 1 plotted as log *I* versus *V*.

Schrieffer and Wilkins where both quasiparticles from the pair in 1 tunnel into 2. Conservation of energy requires that this process has an onset at Δ. For completeness we show in (C) the Josephson current which transfers pairs from 1 to 2 without the excitation of quasiparticles in either superconductor.

As pointed out by Schrieffer and Wilkins, the two-particle process, being proportional to T^4 (where T^2 is the tunneling probability), should be weaker than is observed and a distribution of low spots in the oxide was postulated to explain the experimental magnitudes. In view of the problems commonly encountered in making the junction oxide, this does not seem unreasonable. However, the fact that, at least in our junctions,

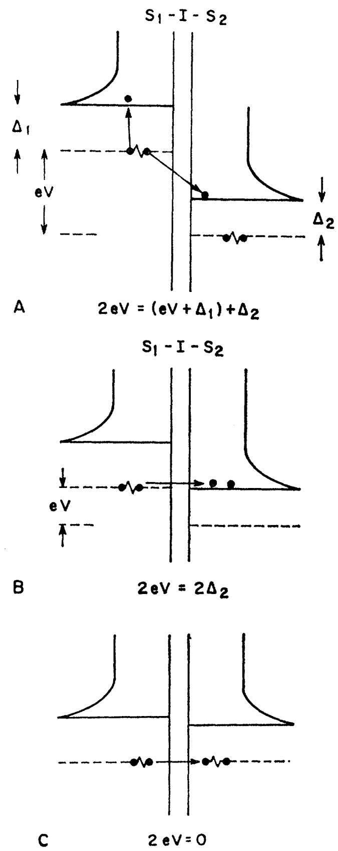

FIG. 4. Representation of the tunneling processes taking place in (A) single-particle tunneling, (B) multiparticle tunneling, (C) Josephson tunneling. The dashed lines represent the Fermi levels and the solid lines the excited particle density of states.

the current onset is not at Δ but at a voltage appreciably less than Δ means the process is not as simple as was assumed. An interesting speculation due to McMillan[18] is that, if a collective mode in superconductors exists as an excited state of the Copper pair, then in order to observe it we must inject two particles into the superconductor. This injection of two particles is the process we believe occurs at Δ in these junctions, so the question of whether the current onset before Δ can be due to excitation of the collective mode seems worth consideration.

IV. SUBHARMONIC TUNNELING—THE *I-V* CHARACTERISTIC

The differences between multiparticle and subharmonic tunneling are obvious when Fig. 1 is contrasted with Fig. 5. The important features of subharmonic tunneling are the following:

(1) The current for $V<2\Delta$ can be an appreciable fraction of that flowing at $V>2\Delta$.

(2) This current for $V<2\Delta$ is independent of temperature (except for changes in position of the structure as Δ changes). In the junction of Fig. 5, even at 4.2°K, this excess current dominates the thermal quasiparticle current.

(3) As discussed by Marcus and to be illustrated below, the current is rather insensitive to a magnetic field.

(4) The structure at Δ is *not* an increase in current but a decrease in conductance.

(5) The current decreases only gradually as V is reduced from 2Δ to zero and structures of a similar shape to that at Δ are observed at $2\Delta/3$, $2\Delta/4$, $\cdots$.

In contrast to Fig. 1, the current does not change by orders of magnitude (or even a factor of 2) near any of these structures. The shape of these $2\Delta/n$ structures seems to be an important clue to the origin of the effect. They can be regarded either as decreases in conductance or, if a smooth background is arbitrarily guessed at, then the structures can possibly be interpreted[7] as peaks in current at $2\Delta/n$. Not by any stretch of the imagination can we claim, however, that the current increases at $2\Delta/n$ as in multiparticle tunneling. Thus it appears that in multiparticle tunneling a new additional tunneling process occurs near Δ; in subharmonic tunneling a process either reaches a maximum or begins to decrease at $2\Delta/n$. Some junctions seem to show both types of behavior; if one examines the *I-V* plots of Taylor and Burstein,[1] one sees the Δ_{Tl} structure is an increase in current and the Δ_{Sn} a decrease in conductance.

The location of the $2\Delta/n$ structure is more easily observed by plotting the derivative of the tunneling characteristic as in Fig. 6. In practice we measure the

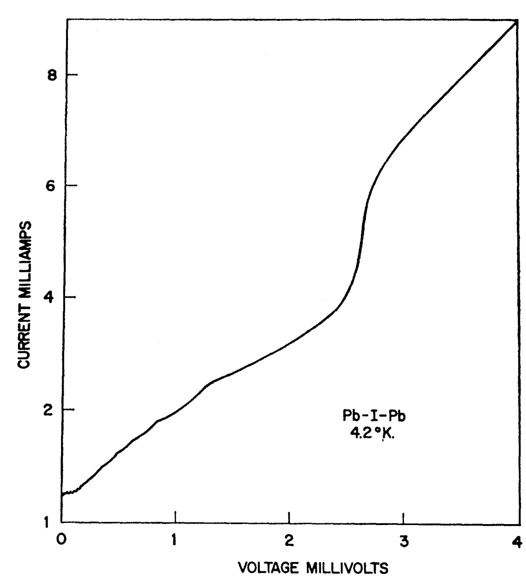

FIG. 5. The *I-V* characteristic of a Pb–*I*–Pb junction at 4.2°K. We consider this to be typical subharmonic junction behavior.

[18] W. L. McMillan (unpublished).

dynamic resistance dV/dI but have plotted this measurement to look roughly like conductance, a useful trick if the resistance changes are small. The minima in conductance are by far the sharpest features of the plot and we take the minimum to measure the position V_n of the $2\Delta/n$ structure. We find that the values of "gap" given by $V_n \times n$ progressively decrease as n increases. In fact, it is often very hard to determine a good gap value from junctions having large subharmonic currents and in the discussion below we have used values given[19] for similar films in junctions of the type in Fig. 3.

V. EVIDENCE FOR SHORTS

Marcus[7] has outlined evidence suggesting the presence of metallic shorts or bridges in junctions which exhibit subharmonic tunneling. Our further evidence is as follows:

(1) We always make five junctions of three different areas[20] at the same time. Often four of these have resistances inversely proportional to junction area to within 20%. The fifth has a resistance much lower than expected. The four "good" junctions generally show multiparticle tunneling. The fifth low-resistance junction shows subharmonic structures.

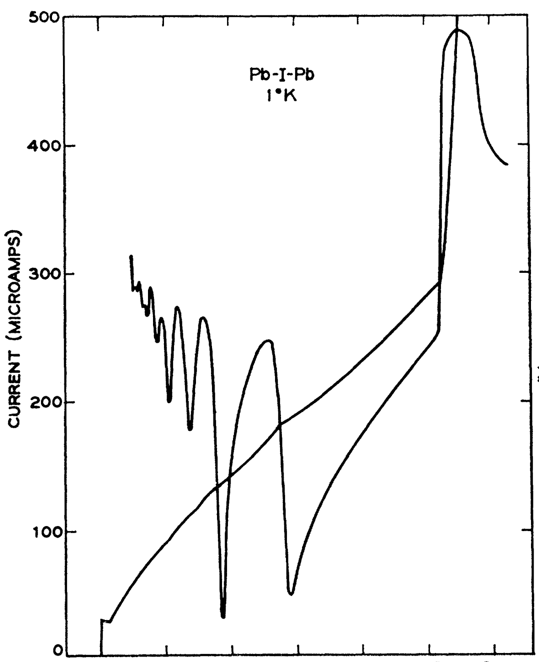

FIG. 6. The I-V characteristic of a "subharmonic" Pb–I–Pb junction at 1°K. We also show the derivative dV/dI (dynamic resistance) plotted downwards, i.e., minima in this plot are minima in *conductance.*

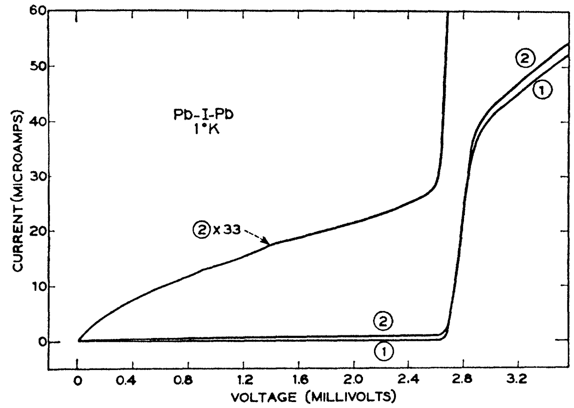

FIG. 7. The I-V characteristics for a Pb–I–Pb junction at 1°K (1) before and (2) after the "accident" discussed in the text.

(2) A junction of the type shown in Fig. 1 was being used for other tunneling studies with remarkable reproducibility over a period of a month or so. One day the junction was lifted out of liquid helium with a rather large applied voltage; on measuring again at 1°K, the I-V characteristic had changed from trace 1 of Fig. 7 to trace 2. When the current is expanded for $V<2\Delta$, trace 1 is similar to Fig. 1, trace 2 is similar to Fig. 5. However, not only had the current increased in the range $V<2\Delta$, but it also increased for all voltages $V>2\Delta$. This strongly indicates that the oxide had been damaged to produce a low spot or short, that this decreased the junction resistance and, in addition, produced the subharmonic behavior.

(3) Believing that the short mentioned above should produce an additional nontunneling path through the junction, we measured the dynamic resistance for $V>2\Delta$ where phonon effects[19,20] produce rather large changes in resistance. In Fig. 8 we compare this measurement for a junction of the type shown in Fig. 1 and one having a subharmonic current. As we expect the phonon effects to produce changes of definite magnitude in the tunneling resistance, the apparent reduction of the effects in the subharmonic junction indicates the presence of a parallel nontunneling path—the short. In fact, the conductance of the short could be deduced from Fig. 8 and gives an extrapolated current for $V<2\Delta$ which is comparable to, but smaller than, the measured excess current in that region.

(4) The I-V trace for an S–I–S junction in the superconducting state should approach that in the normal state at high voltages. This normal-state trace can be obtained by raising the temperature or applying a magnetic field. On applying a large magnetic field to a junction which showed an appreciable subharmonic current, we observed (Fig. 9) that the S–I–S trace increased above the M–I–M trace near 2Δ and,

[19] W. L. McMillan and J. M. Rowell, Treatise on Superconductivity, edited by R. Parks (Marcel Dekker, New York, to be published).

[20] J. M. Rowell and L. Kopf, Phys. Rev. **137**, A907 (1965).

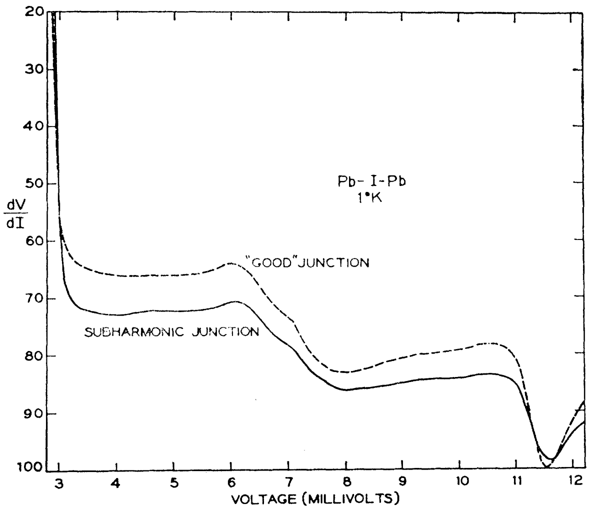

FIG. 8. The resistance dV/dI versus V plots for two Pb–I–Pb junctions at 1°K. The solid line is for a junction with subharmonic effects, the dashed line for a "good" junction similar to Fig. 1.

in fact, exceeded this normal current until $V\sim 70$ mV for this particular junction. This was immediately suggestive of bridge behavior,[21] where the superconducting characteristic exceeds the normal at all low voltages. In fact, the voltages where the two traces coincide for a bridge structure do not seem to be mentioned in the literature.

(5) Eventually, by chance, the limiting case of subharmonic behavior was observed and is shown in Fig. 10. In this "junction" there is *no* evidence of single-particle tunneling as the structure at 2Δ is also a decrease in conductance corresponding exactly in shape

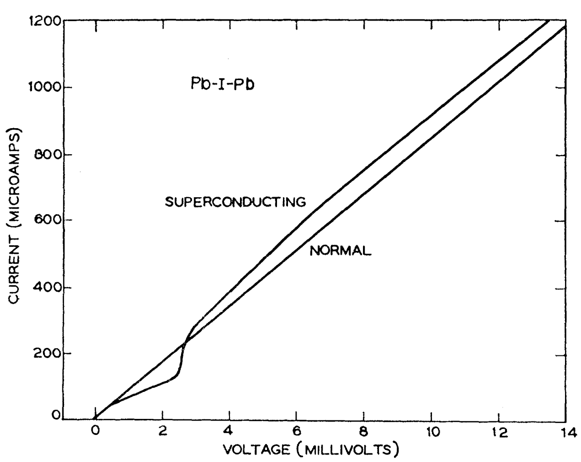

FIG. 9. I-V characteristics for a Pb–I–Pb junction at 1°K in the superconducting and normal states. The normal state was produced by application of a magnetic field.

[21] P. W. Anderson and A. H. Dayem, Phys. Rev. Letters **13**, 195 (1964); A. H. Dayem and J. J. Wiegand, Phys. Rev. **155**, 419 (1967).

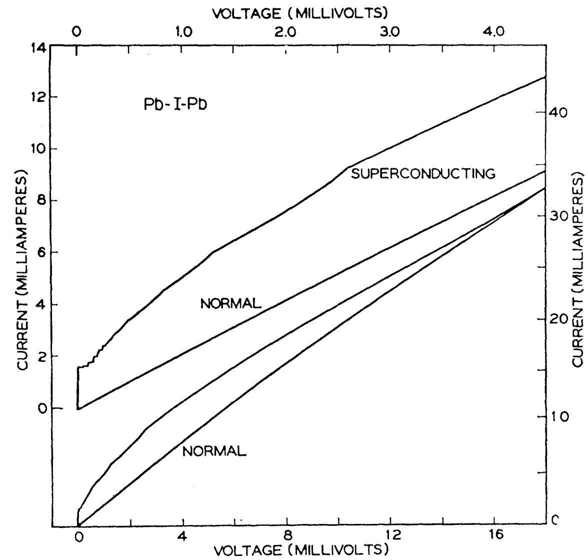

FIG. 10. Normal and superconducting I-V characteristics for a Pb–I–Pb junction at 1°K which exhibits no single-particle tunneling. The upper traces use the left and top scales, the lower traces the right and bottom scales.

to those at $2\Delta/n$. In this case the superconducting trace exceeded the normal for all voltages up to 17 mV.

The shape of the 2Δ structure is seen more clearly in Fig. 11, where the derivative plot shows the decrease in conductance near $2\Delta_{Pb}$ and also the decreases at $2\Delta/n$ which become progressively sharper as n increases.

Thus we claim that the subharmonic behavior is probably not a tunneling process, that it is independent of any single-particle tunneling that may be occurring in the junction, that the characteristic exhibits

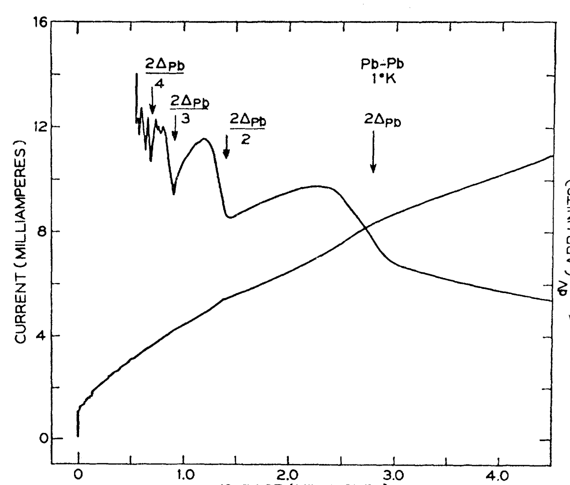

FIG. 11. Current and dV/dI versus voltage for a Pb–I–Pb junction at 1°K which shows no single-particle tunneling. Note the *decrease* in conductance at 2Δ.

decreases in conductance at 2Δ and $2\Delta/n$, and that the structure arises from the presence of a short or bridge in the junction. Whether it is in fact necessary that the bridge be surrounded by the cavity of the junction is not clear; perhaps Anderson-Dayem bridges could exhibit $2\Delta/n$ structures under certain conditions.

VI. MODELS TO EXPLAIN SUBHARMONIC BEHAVIOR

Although, as we will show later, we do not believe the correct explanation of the subharmonic characteristic has been proposed, we will outline a few of the possibilities to show what mechanisms have been considered. Although many of these possibilities have been discussed privately for some time, they were first considered seriously by Werthamer[22] and conveniently reviewed by Rochlin.[8] It has been assumed that in the junction we have a single-particle tunneling current and a Josephson current giving rise to microwave power of frequency $2eV/h$. Although some of the junctions shown in this paper did exhibit Josephson currents (quenched by a magnetic field when necessary in order to show the low-voltage I-V characteristic), the shorts we claim exist will give power at $2eV$ and also in the harmonics $n\times 2eV$,[21] so it is not necessary for the junction to be of a low enough resistance to have a Josephson current. This ac field can interact with the junction in the following ways (and as the ac power is fed from the external dc source, any abrupt changes in the interaction will be reflected in the dc I-V characteristic):

(1) The ac field can be absorbed by the films of the junction, creating quasiparticles in each film, as shown in Fig. 12. This occurs at $n\times 2eV\geq 2e\Delta_1$ and $n\times 2eV\geq 2e\Delta_2$ (we will consider an asymmetrical junction S_1–I–S_2) or $V=\Delta_1/n$, Δ_2/n. This is the even series $2\Delta/2n$ which appears in the calculation of Werthamer; as the absorption of ac power increases, we might expect the I-V characteristic to show an increase in current near these voltages also.

(2) The radiation can interact with the single-particle tunneling current in the way described by Dayem and Martin[23] for an externally applied field. In their case they observed current steps at $eV=(\Delta_1+\Delta_2)-nh\nu$, where ν was their applied frequency. In the Josephson junction, as suggested to the author in 1963 by Anderson, interactions should occur at $eV=(\Delta_1+\Delta_2)-n\times 2eV$ or at $V=(\Delta_1+\Delta_2)/(2n+1)$—this has come to be called the odd series. This argument can be extended so that the interaction is with the two-particle current at $\Delta_1/(n+1)$, $\Delta_2/(n+1)$ but it seems impossible that this process could have a measurable magnitude.

[22] N. R. Werthamer, Phys. Rev. **147**, 255 (1966).

[23] A. H. Dayem and R. J. Martin, Phys. Rev. Letters **8**, 246 (1962).

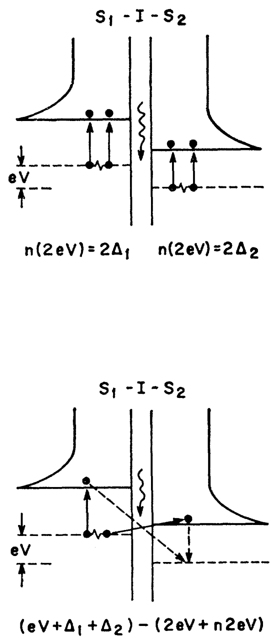

FIG. 12. The upper diagram represents the absorption of Josephson radiation by the superconducting metal films comprising the junction. The lower diagram represents the intermediate tunneling state of the Josephson ac current and the interaction of radiation with this intermediate state.

(3) A third possibility arises from the work of Riedel,[24] who showed that the amplitude of the ac Josephson current has a singular peak at $eV=2\Delta$ or for a frequency of 4Δ. This rather surprising result has also been discussed by Werthamer[22] and by Anderson.[25] They show that the amplitude of the pair current varies with voltage and the expression for this amplitude has an energy denominator which is the energy of the intermediate tunneling state. This intermediate state (Fig.12) is due to the destruction of a pair in S_1, the excitation of one quasiparticle in S_1 and the other in S_2—exactly as in single-particle tunneling. The transfer then proceeds as the quasiparticle in S_1 tunnels to S_2 and recombines with that in S_2 as a pair at the Fermi level. For an applied bias V the energy of this intermediate state is $(eV+\Delta_1+\Delta_2)-2eV=\Delta_1+\Delta_2-eV$. Thus we expect the pair amplitude to peak when this is a minimum, at $eV=2\Delta$ for a symmetrical junction and at $eV=\Delta_1+\Delta_2$ for an asymmetrical junction. If we extend this argument so that a field of frequency $n\times 2eV$ exists in the junction, then the intermediate state can be reached with the help of absorption from this field and its energy is

$$(eV+\Delta_1+\Delta_2)-(2eV+n\times 2eV)=\Delta_1+\Delta_2-(eV+n\times 2eV)$$

and the amplitude maxima would be at

$$(\Delta_1+\Delta_2)/(2n+1).$$

[24] E. Riedel, Z. Naturforsch. **19A**, 1634 (1964).

[25] P. W. Anderson, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland Publishing Co., Amsterdam, 1967), Vol. 5. See, in particular, pp. 7–9, Eq. (2.12′).

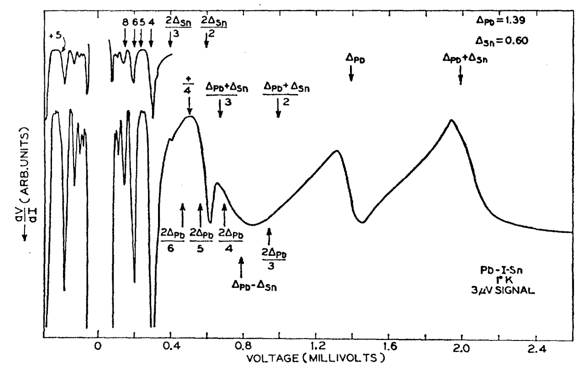

FIG. 13. Dynamic resistance dV/dI versus voltage for a Pb–I–Pb (lead-oxide insulator) junction at 1°K. The modulating current gave a 3μV full-scale voltage for the lower trace; the current was reduced for the upper trace. The positions of possible subharmonic series are indicated. $+/4$ means $\Delta_{Pb}+\Delta_{Sn}/4$; 4, 5, etc., at the top of the figure mean $2\Delta_{Sn}/4$, $2\Delta_{Sn}/5$, etc.

The above summarizes very simply the interactions of the Josephson ac field with the junction that have been proposed to date. We see that, in order to explain the observed $2\Delta/n$ series, we have to invoke two mechanisms to give an odd and an even series. As, experimentally, the shapes of the odd and even structures appear identical and their magnitudes are not appreciably different, it appears that a single explanation of the whole series is yet to be found.

VII. TUNNELING IN ASYMMETRICAL JUNCTIONS

All the results on subharmonic tunneling published to date were obtained for symmetrical junctions. It was suggested by Werthamer[22] that a study of asymmetrical S_1–I–S_2 junctions would be useful, and we have performed measurements of Pb–I–Sn (lead-oxide insulator), Sn–I–Pb (tin-oxide insulator), and Sn–I–In junctions. The results for Pb–I–Sn and Sn–I–Pb junctions are the same and a typical derivative plot for a Pb–I–Sn junction is shown in Fig. 13. The following points should be noted:

(1) At $\Delta_{Pb}+\Delta_{Sn}$, the structure is a mixture of a decrease in conductance and a hint of the single-particle peak. It appears that very little single-particle tunneling is occurring in this junction.

(2) Structure is observed at Δ_{Pb} but not at $2\Delta_{Pb}/n$ for $n>2$ with n even or odd.

(3) The series $(\Delta_{Pb}+\Delta_{Sn})/n$ with n even or odd is not observed.

(4) The dominant series observed is $2\Delta_{Sn}/n$ with n even. This series with n odd is not observed, except for possibly a very weak structure at $2\Delta_{Sn}/3$.

Considering the explanations in Sec. VI, we see that:

(1) If absorption in the films is occurring (at $2\Delta/2n$), it is only in the tin film, as the series $2\Delta_{Pb}/2n$ is not observed for $n>1$. As the tin gap is smaller, it would require a lower harmonic to excite tin quasiparticles than lead quasiparticles at any given voltage $<\Delta_{Sn}$.

(2) Interaction with single-particle tunneling at $(\Delta_{Pb}+\Delta_{Sn})/(2n+1)$ is not taking place. This is not surprising as we have shown that subharmonic structure can be observed when there is no single-particle tunneling in the junction.

(3) The Riedel peaks at $(\Delta_1+\Delta_2)/(2n+1)$ do not occur.

We found that the behavior shown in Fig. 13 is typical of both Pb–I–Sn and Sn–I–Pb junctions, the insulator being the oxide of the first metal, for example, lead oxide in Pb–I–Sn junctions. In order to investigate an asymmetrical junction of a different type, Sn–I–In structures were made without realizing that, at low temperatures, Δ_{Sn} and Δ_{In} are practically identical. Unfortunately the type of junction where the bridge carries all the current did not occur in the investigation of this system and the thermally excited current obscured the excess current at higher temperatures where $\Delta_{Sn}\neq\Delta_{In}$. At low temperatures the I-V characteristic gave $\Delta_{Sn}+\Delta_{In}=1.235$ mV and Fig. 14 shows that these junctions exhibited a multiparticle peak in conductance at $(\Delta_{Sn}+\Delta_{In})/2$. Below this voltage the characteristic subharmonic structures are observed at $(\Delta_{Sn}+\Delta_{In})/n$, with $n=3$, 4, 5. At the lowest voltages the structures for $n>5$ seem to be washed out, in contrast to all the other junctions described above where the highest-order structures are the sharpest.

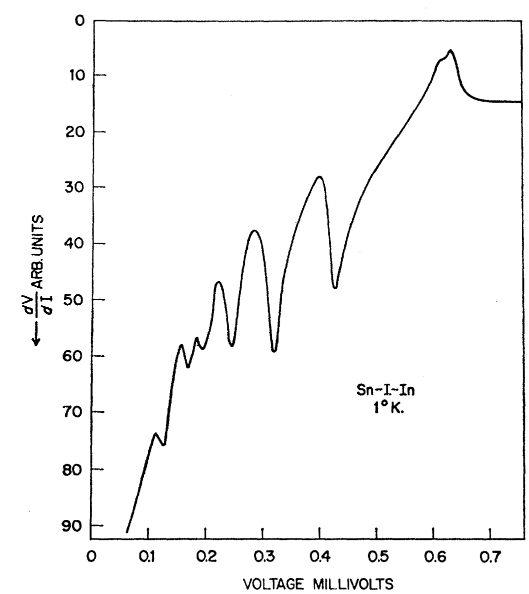

FIG. 14. dV/dI versus V for a Sn–I–In junction at 1°K. The value for $\Delta_{Sn}+\Delta_{In}$ obtained from the I-V characteristic is 1.235 mV.

It has been pointed out to us by Shen that Al–I–Al junctions showed no multiparticle tunneling or subharmonic behavior[1] and it is our experience that in Al–I–M junctions (M=Pb, Sn, In) subharmonic behavior is not observed. This is presumably because of the much greater perfection of the aluminum-oxide layer (this oxide is produced with little difficulty compared to lead and tin oxides) and the resulting absence of shorts.

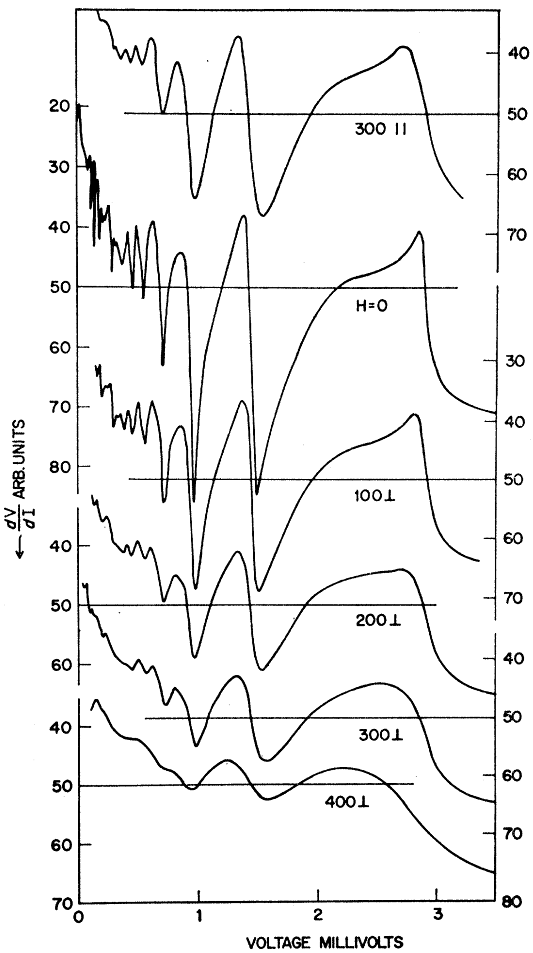

FIG. 15. The effect of magnetic field on the dynamic-resistance-versus-voltage plots for a Pb–I–Pb junction at 1°K. The resistance scales are offset for clarity and a line is drawn at $dV/dI=50$ for each field. Reading from the top, the fields are 300 G parallel to the plane of the junction and 0, 100, 200, 300, and 400 G perpendicular to the junction.

The only conclusion that can be drawn from these measurements of asymmetrical junctions is that there is no satisfactory explanation of the behavior of subharmonic tunneling in asymmetrical and symmetrical junctions. Even if, by invoking two separate mechanisms, one considers the symmetrical junction to be understood, then the reason why these two mechanisms do not operate in the asymmetrical junction remains unexplained.

VIII. EFFECT OF MAGNETIC FIELD

The subharmonic-tunneling structure is relatively insensitive to field, as reported by Marcus.[7] This seems to eliminate any interactions of the ac field with particular resonances of the junction cavity. The derivative trace as a function of field is shown in Fig. 15. It can be seen that 300 G parallel to the junction has very little effect, while 400 G transverse almost washes out the structure. This suggests that the effect of the field is simply to introduce normal regions into the films (at 400 G approximately $\frac{1}{4}$ of the film will be normal) and thus different areas in the junction will have gap values anywhere from zero to Δ.

IX. CONCLUSION

The study of asymmetrical tunnel junctions, which was undertaken in an attempt to clarify our understanding of the symmetrical junction, has led us to the conclusion that the interaction responsible for subharmonic tunneling structure is not understood at all. It seems reasonable that it is due to the interaction between the ac field from a metallic bridge and the films and/or cavity of the junction. As we do not believe that single-particle tunneling is necessary to the observation of the effect, it might be worthwhile to study bridges of the Anderson-Dayem type (possibly surrounded by, but electrically insulated from, superconducting material) or nontunneling point contacts in order to see whether similar structures can be observed there.

ACKNOWLEDGMENTS

We wish to thank L. Kopf for technical assistance during part of this research and we are indebted to P. W. Anderson, W. L. McMillan, and N. R. Werthamer for a number of valuable discussions.