Subharmonic Structure in Superconducting Tunneling

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(Received 3 November 1969)

The current-voltage characteristic obtained in a tunneling experiment between two equal superconductors often contains structure at $2\Delta/n$ where 2Δ is the energy gap and n is an integer. The origin of this subharmonic structure has never been satisfactorily established; however, it has often been linked with the Josephson effect. We have studied this subharmonic structure experimentally and have obtained three new results: (1) The magnitude of the structure can be made to increase proportionally to the square of the tunneling matrix element; (2) the structure is very sensitive to externally applied microwave radiation; (3) when two dissimilar superconductors a and b are used, the structure appears at $2\Delta_a/2n$, $2\Delta_b/2n$, and $(\Delta_a+\Delta_b)/(2n+1)$. These experimental results strongly support the idea that the structure is caused by the self-detection of the Josephson radiation. To explain the magnitude of the structure, it is necessary to assume that the tunneling matrix element is so large that it is improper to linearize the Josephson equations.

INTRODUCTION

The current-voltage characteristic of a superconducting tunnel junction can be calculated using the simple BCS theory,[1] and it is possible to fabricate tunnel junctions which adhere very closely to the calculated characteristics.[2] However, very often the observed current-voltage characteristics deviate markedly from theory. One of the first anomalies was reported by Taylor and Burstein[3] who noticed a small onset in the current when the applied voltage was equal to half the energy gap, Δ/e, in a tunneling experiment between two equal superconductors. Schrieffer and Wilkins[4] attempted an explanation where they noticed that if two electrons could tunnel simultaneously, this process would become energetically possible at a voltage corresponding to half the energy gap, and cause a structure in the current-voltage characteristic akin to the observed anomaly. The difficulty with this explanation is that the observed anomaly should depend on the fourth order of the tunneling matrix element M and therefore should be practically unobservable. This objection was circumvented by assuming that the junction contained thin spots where the matrix element would be much larger and that these thin spots would be responsible for the anomalous current flow.

Relatively soon afterwards it was apparent[5] that not only is there an anomaly in the current at a voltage corresponding to half the energy gap, but in fact at all submultiples of 2Δ, i.e., at $2\Delta/n$. Here n is an integer and the tunneling experiments were performed between two identical superconductors. The present situation has been discussed recently by Rowell and Feldmann.[6] Using a suggestion by Werthamer[7] they also investigated the subharmonic structure in junctions made of dissimilar superconductors; however, their conclusions are in conflict with some of our results, which will be reported in this paper. Werthamer suggested that the subharmonic structure could be caused by a self-detection of the ac Josephson effect.[8] We feel strongly that our results substantiate his views, and that indeed some of the excess current, and in particular, the subharmonic structure in a tunnel junction is due to the Josephson effect. We hope to demonstrate that essentially only one mechanism is responsible for the observed structure, in contrast to the classification attempted by Marcus[9] and which was discussed in some detail by Rowell and Feldmann.[6] In particular we do not believe that "the multiparticle tunneling" discussed in detail by Wilkins[10] has ever been observed.

EXPERIMENTAL RESULTS

A. Experiments with Evaporated Barriers

Normally, superconducting tunnel junctions are made of thin evaporated-metal films which are spaced apart by a naturally grown oxide layer.[2] Recently it was discovered that it is possible to make tunnel junctions where the barrier consists of an evaporated semiconductor.[11] This is particularly interesting when a light-sensitive semiconductor such as CdS is used, because we can then adjust the strength of the matrix element in the junctions, as discussed by us in an earlier paper.[12] In this case it will allow us to determine the relationship between the appearance of the subharmonic structure and the strength of the matrix element.

The sample preparation is pictured in Fig. 1. First a metal such as Sn is evaporated onto a glass slide in the form of a narrow strip. Im-

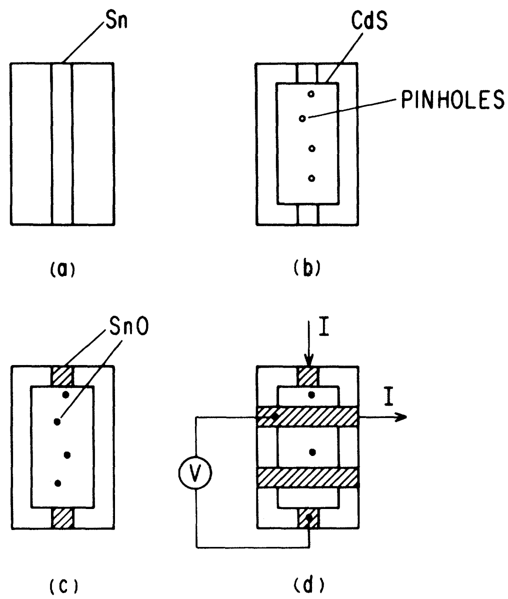

FIG. 1. Sample preparation. (a) An evaporated Sn film on a glass slide. (b) A CdS film has been deposited on top of the Sn film. Experience shows that the CdS film will not be continuous but will contain small pinholes. (c) The sample is exposed to air and an insulating layer of SnO is formed inside the pinholes. (d) A cross strip of Sn is evaporated to complete the junction.

mediately and without breaking the vacuum, a thin layer of CdS is evaporated on top of the Sn film. Experience shows that the CdS layer is not continuous but will contain small pinholes. Next, the assembly is exposed to air, and where the Sn is exposed through a pinhole in the CdS layer, it will oxidize. After a suitable time the assembly is put back into the evaporator, and, to complete the tunnel junction, cross strips of any metal, for example, Sn, are deposited over the CdS layer. In its final form the two Sn strips are separated from each other either by the CdS layer or by the Sn oxide where a pinhole existed. A schematic cross section is shown in Fig 2.

When a voltage is applied between the two Sn strips, the electrons can flow either through the CdS layer itself which covers most of the area, or through the SnO inside one of the small pinholes. By varying the oxidation time, either of these two contributions to the total current can be made to dominate.

A typical experimental result is shown in Fig. 3. Before the sample has been exposed to light, we have almost an ideal single-particle tunneling characteristic. After the sample has been exposed to light, we see that the current-voltage characteristic has changed appreciably and that a dc Josephson current is observable. We also see a rather large structure in the current-voltage characteristic at an applied voltage equal to half the energy gap. By exposing the sample to light, the

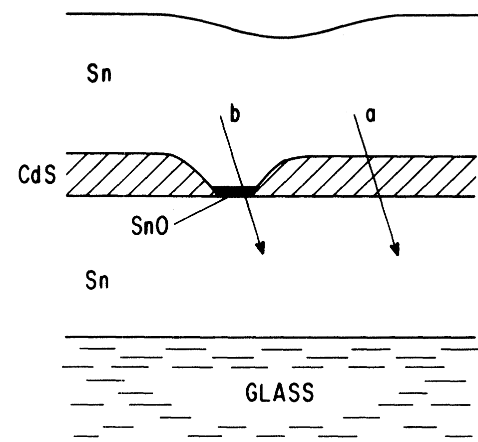

FIG. 2. Schematic cross section of a junction through one of the pinholes. Electrons can flow between the two Sn films either through the CdS layer indicated by arrow a or through the SnO indicated by arrow b. Either of these two currents may dominate the current-voltage characteristic depending mainly on the relative thickness of the SnO and the CdS layer.

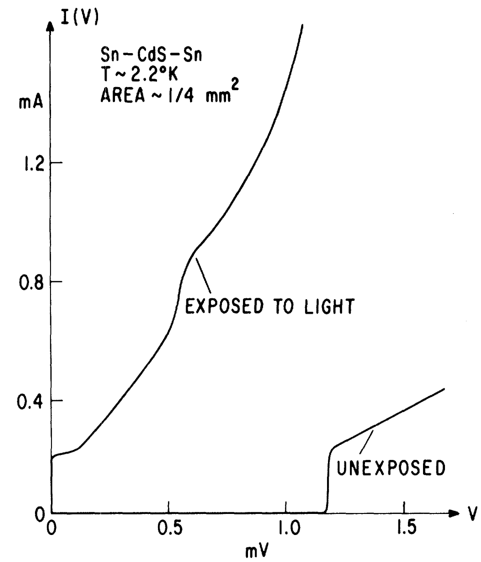

FIG. 3. Current-voltage characteristic of a Sn-CdS-Sn junction before and after the junction has been exposed to light. In the unexposed state an almost ideal single-particle tunneling characteristic is obtained, while in the exposed state a dc Josephson current and a large mid-gap structure are readily visible. The junction has been shielded from the earth's magnetic field.

tunneling matrix element has increased, accounting for the gross changes in the current-voltage curve. Note that it is enough to expose the sample to light; the changes remain after the light has been shut off. Such an infinite-lifetime effect is previously known for CdS and other II–VI compounds and is commonly called conductivity storage.[13–15] The original resistive state of the junction can be recovered by briefly heating the junction to above 110°K and recooling. It must be noted here that not all the current which flows through such a sample is due to electrons tunneling directly from one superconductor to the other. Apparently a substantial fraction of the electrons can tunnel into an intermediate state in the barrier, as discussed in an earlier paper.[12] We shall here only be concerned with the part of the excess current which causes the subharmonic structure and thus neglect all other possible forms of excess current.

The subharmonic structure we obtain with the CdS junctions has a different appearance depending on whether the current through the oxide-covered thin spots dominates the current-voltage characteristic or the current through the CdS layer itself dominates. The former gives rise to a cusplike structure in the current-voltage characteristic at $2\Delta/n$, while the latter gives rather a steplike increase in the current at these values. Rowell and Feldmann labeled these two behaviors as subharmonic structure and multiparticle structure, respectively; however, we believe that only one mechanism is responsible for the behavior. In the following we shall give examples of the two extreme cases. It should be understood that a large amount of the data which we have collected falls in between these two cases.

Figure 4 shows a plot of the current-voltage characteristic of a Sn-CdS-Sn sample prepared as described above where the current through the CdS dominates the characteristic. The plot is on a semilog paper and the change in the square of the tunneling matrix element, M^2, is approximately a factor of 2 between each curve. This increased current is accomplished by exposing the sample to light for a brief period between the experiments represented by each curve. As is seen, the virgin curve does not contain a visible half-gap structure. Initially the size of the structure at the half-gap grows more rapidly than the tunneling current but soon saturates and grows at the same rate as the tunneling current. Thus we may conclude that the structure at half-gap increases faster than M^2 for a weakly coupled system and proportional to M^2 for a strongly coupled system.

This is true not only for the half-gap structure but also for the whole subharmonic series as

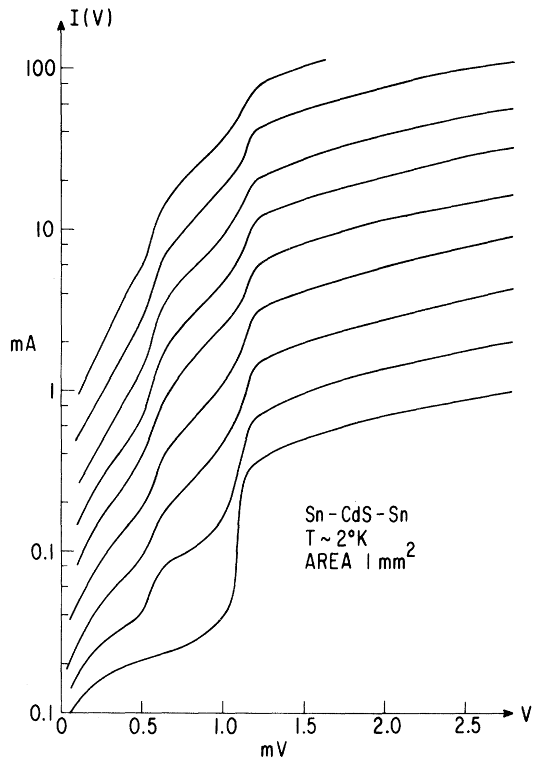

FIG. 4. Current-voltage characteristic of a Sn-CdS-Sn junction plotted on semilog paper. Each curve represents a separate run, and the junction has been exposed to light for a brief period between each experiment. Note that the current contributing to the mid-gap structure increases faster than the total current at first, but soon saturates and increases proportionally to the total current.

shown in Figs. 5(a) and 5(b) which plot the current-voltage characteristic and the derivative dV/dI versus voltage, respectively, for a Pb-CdS-Pb sample for various values of the matrix element. Thus it appears that all the excess current which causes the subharmonic structure can be made proportional to the tunneling matrix element squared.

Figure 6 shows an example of a junction where the current is dominated by the thin spots in the sample. The sample was only briefly exposed to air after the evaporation of the CdS layer such that the pinholes would be covered only by a very thin oxide. Note that the structure at the energy gap itself is barely visible in the dark state. In many aspects this sample is similar to one of the results reported by Rowell and Feldmann where they believed that they had a metallic short. By changing the matrix element by exposing the sample to light, the ordinary-gap structure as well as the half-gap structure becomes much more prom-

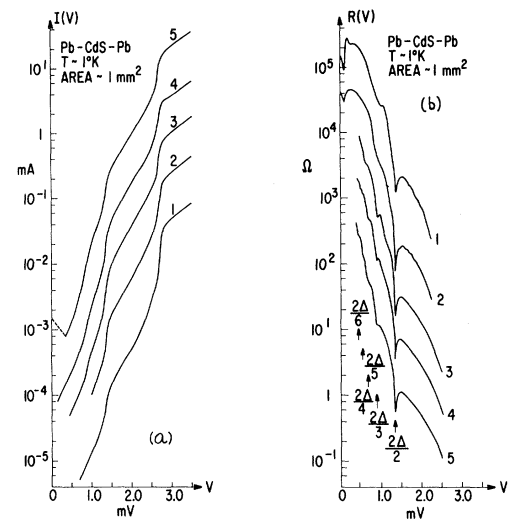

FIG. 5. (a) Current-voltage characteristic of a Pb-CdS-Pb junction for various values of the tunneling matrix element. (b) Dynamic resistance dV/dI versus voltage of the same junction displayed in (a). Note that all the subharmonic structure increases linearly with the total current.

inent. Initially all the current flows through one of the thin spots in the CdS where a finger of Sn protrudes into the CdS, but is separated from the other Sn layer by an extremely thin oxide. When light is applied some of the current can tunnel through the CdS as well. Even though the appearance of the subharmonic structure is somewhat different before and after the junction has been exposed to light, we believe that the structures in both cases are caused by the Josephson effect. The cusp structure which extends to a large n in the series $2\Delta/n$ is present when the current through the junction is dominated by a thin spot where the matrix element is very large, while the more smeared steplike structure is present when the current through the total area dominates and the matrix element is smaller.

The thin-spot or "finger" theory is substantiated by experiments where Ge is used as the evaporated layer. The current-voltage characteristic shown in Fig. 7 is readily obtained if the sample is prepared as follows: A Pb strip is evaporated onto a glass slide, oxidized for a few minutes, and then covered with a thin layer of Ge and finally with cross strips of Pb. If the short oxidation time is omitted, the sample is invariably shorted. By applying a larger voltage to these samples the cur-

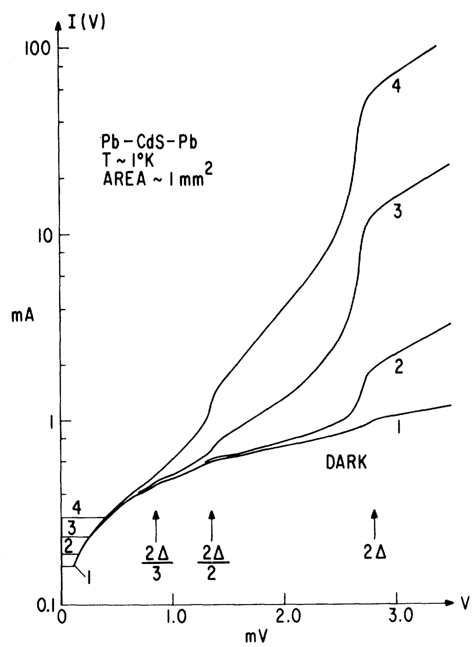

FIG. 6. Current-voltage characteristic of a junction dominated by thin spots. As the junction is exposed to more and more light, more and more of the total current flows through the CdS layer. Most of our junctions fall between the cases displayed in Fig. 4 and the case shown in this figure.

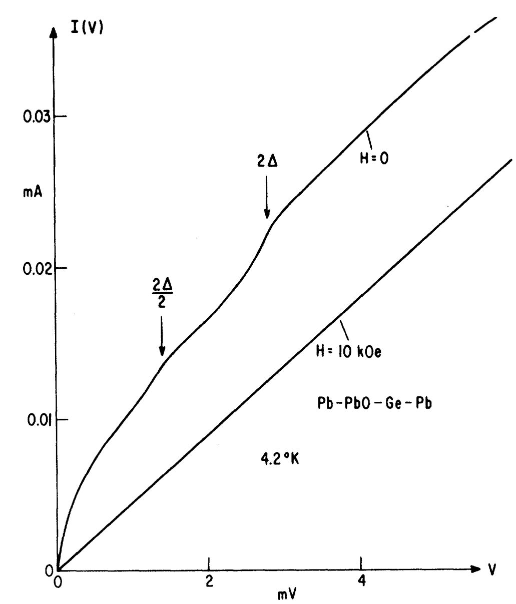

FIG. 7. Pb-PbO-Ge-Pb junction dominated by thin spots. Because of the very large coupling between the two Pb films at a thin spot, the Josephson ac effect dominates the single-particle tunneling, causing the unfamiliar appearance of the curve. We do not believe that a direct metallic short is responsible for this behavior.

rent-voltage curve shows a certain heating effect reported elsewhere,[16] which substantiates the finger theory.

A consequence is that the subharmonic structure seen in the current-voltage curve is sometimes associated with only one of these fingers. For various reasons, such as heating or large current density, these fingers may have an energy gap slightly smaller than the bulk value. An example of this is shown in Fig. 8, where the harmonic structure is clearly associated with a smaller energy gap than the main gap, and both the gaps can be seen in the curve. Exposure of the sample to light affects only the structure at the main gap but neither the structure at the gap due to the finger nor its associated subharmonic structure. If several fingers, each with a different gap, are involved a large smearing effect will be present in the subharmonic structure. We believe that the origin of the two gaps in Fig. 8 is due to simple effects such as stated, rather than to an intrinsically anisotropic energy gap postulated by Rochlin.[17]

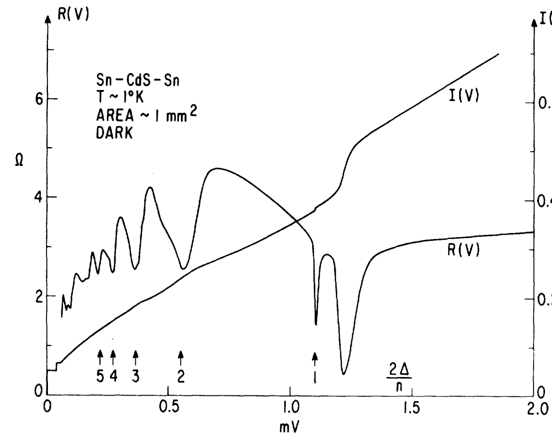

FIG. 8. Current-voltage and dynamic resistance-voltage characteristics of a Sn-CdS-Sn junction. Note that two gaps are visible and that the subharmonic structure can be associated with the smaller gap.

B. Experiment with Microwave Radiation

The experiments with CdS indicated to us that the subharmonic structure somehow is connected with the Josephson radiation; therefore we decided to expose a sample containing such structure to an external source of microwave radiation. If the subharmonic structure is indeed caused by a self-detection of Josephson radiation, we would expect that the structure should also depend sensitively on applied radiation. We actually used as an external source another tunnel junction exhibiting a large Josephson radiation, and this radiation was coupled into our sample in a manner described earlier.[18] The typical results are that the radiation has a large effect on the subharmonic structure, and that the structure in general is smeared by the radiation as shown in Fig. 9. Note in Fig. 9 that we see the induced Josephson current close to zero voltage as was first reported by Shapiro[19] and the induced single-particle tunneling at a voltage close to 2Δ as was first reported by Dayem and Martin.[20] A more remarkable example is shown in Fig. 10. We see that the simple-midgap structure at $2\Delta/2$ has been split into two struc-

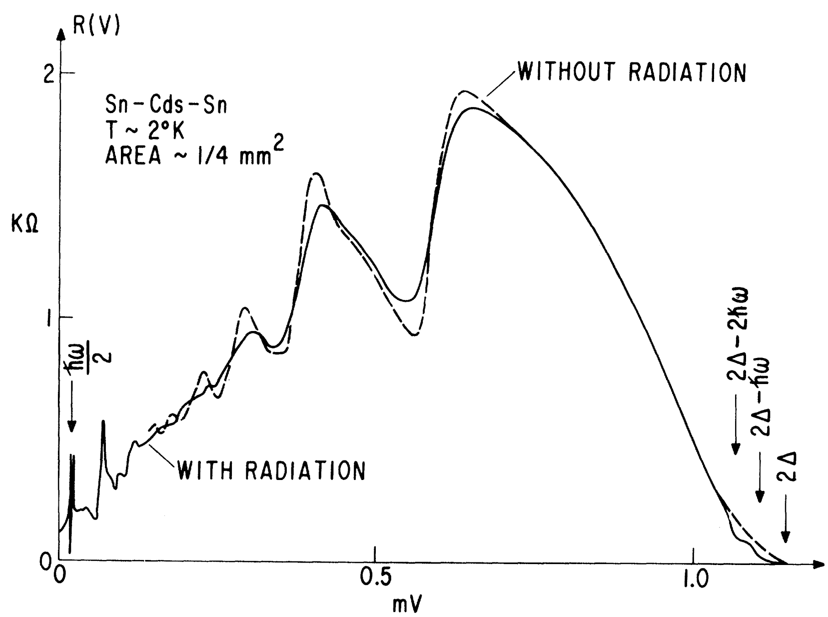

FIG. 9. Dynamic resistance dV/dI as a function of voltage for a junction both with and without an external radiation of energy $\hbar\omega$ applied to the junction. Note the induced pair tunneling near zero voltage and the induced single-particle tunneling near a voltage equivalent to 2Δ. The radiation has a pronounced effect on the subharmonic structure.

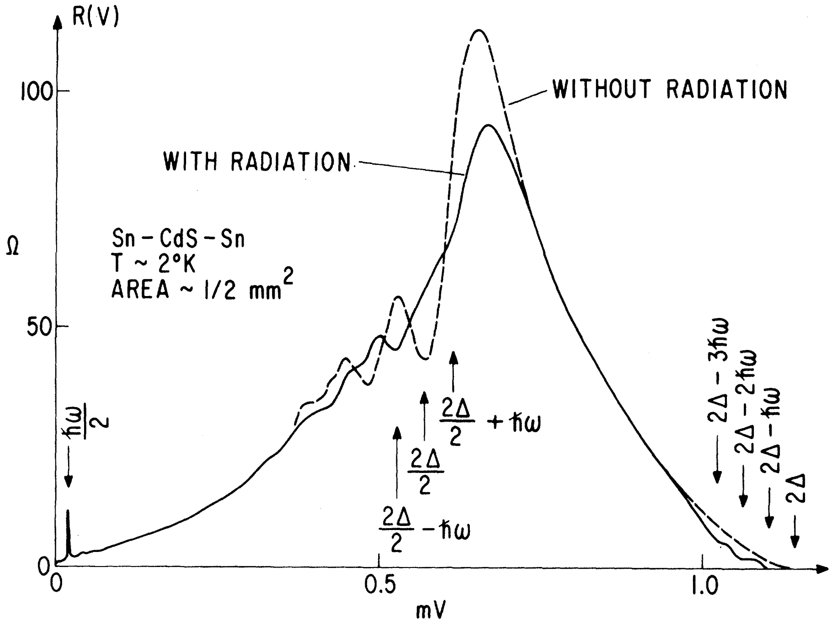

FIG. 10. Dynamic resistance dV/dI as a function of voltage for a junction both with and without an external radiation of energy $\hbar\omega$ applied to the junction. Note that the applied radiation appears to split the midgap bump into two structures.

tures at $2\Delta/2-\hbar\omega$ and $2\Delta/2+\hbar\omega$. This is a somewhat surprising result which we will discuss in more detail later.

In all these cases it is important to keep the radiative coupling into the junction sufficiently weak; otherwise the Dayem-Martin effect will completely dominate the results. This is shown in Fig. 11. We see that because of the strong coupling it is difficult to draw any clear conclusions from Fig. 11. We emphasize that a certain selection of the data has been made to illustrate our experimental findings. Since the subharmonic structures in two different junctions never look exactly similar, selection is, of course, necessary. However, because it is possible to obtain such simple experimental results as shown in Figs. 9 and 10, we are willing to call them "typical samples" and base our conclusions on these results.

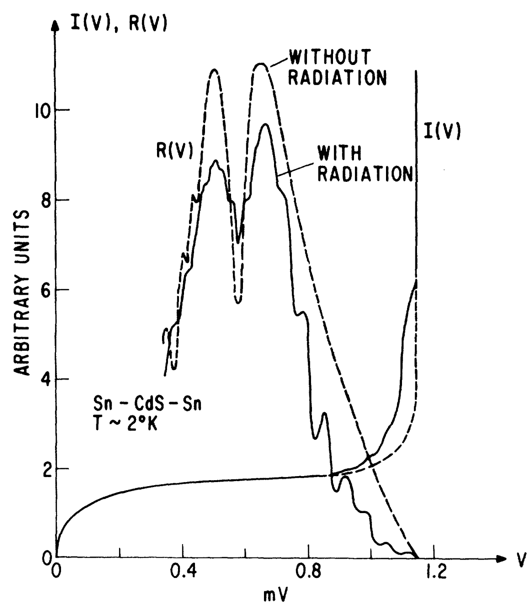

FIG. 11. Current-voltage and dynamic resistance-voltage characteristics for a junction with and without radiation applied. Because of the strong coupling to the radiation field, the induced single-particle tunneling dominates the whole curve.

C. Experiments with Dissimilar Junctions

The effect of radiation on the harmonic structure again suggests that the Josephson effect is responsible for the effect. Thus we decided to follow up an old suggestion of Werthamer[7] and to use two different superconductors to study the odd and even subharmonic structure. Werthamer suspected that if two dissimilar superconductors such as Sn and Pb are used to form a junction, the even structure should occur at $2\Delta_{Sn}/2n$ and $2\Delta_{Pb}/2n$, and the odd structure at $(\Delta_{Sn}+\Delta_{Pb})/(2n+1)$ where n is an integer. This prediction is based upon the assumption that the Josephson effect is responsible for the subharmonic structure. Such experiments have already been reported by Rowell and Feldmann,[6] who, in contrast to us, do not confirm the prediction. Typically, our results on Pb and Sn junctions look similar to theirs, where the $2\Delta_{Sn}/2n$ is the dominant series. However, we can also make junctions where the $2\Delta_{Pb}/2n$ series is apparent, as well as some of the members of the series $(\Delta_{Pb}+\Delta_{Sn})/(2n+1)$. A good result is shown in Fig. 12 for a Pb-Ge-Sn junction which is strongly coupled. Even more significant than the peaks which can be observed are the peaks which are clearly absent such as $\frac{1}{2}(\Delta_{Sn}+\Delta_{Pb})$ or $\frac{1}{3}2\Delta_{Pb}$. At low voltages the assignment of peaks becomes very complex and probably meaningless because of the Fiske steps induced by the Josephson current.[21] The difficulty in observing all these series is that numerically some of them overlap. For example, Rowell and Feldman observed a weak structure at $\frac{1}{3}\times 2\Delta_{Sn}$, while we believe this structure belongs instead to $\frac{1}{5}(\Delta_{Pb}+\Delta_{Sn})$ which is numerically about the same.

In Fig. 13 is shown the result for a strongly coupled Pb-Ge-Tl junction, and this result is somewhat similar to that of Rowell and Feldman on Pb-PbO-Sn. The even structure related to the energy gap in Tl and Pb is clearly visible, while it is difficult to identify odd structure due to $(\Delta_{Pb}+\Delta_{Tl})/(2n+1)$.

If the structure indeed is due to Josephson radiation, it is clear why the even structure caused by the smaller gap is dominating. For small applied voltages excitations can only be formed in the superconductor with the smaller gap, while when the voltage is large enough to cause excitations in the larger gap superconductor, a large amount of the radiation will still be absorbed in the superconductor with the smaller gap. This is true for the absorption related to the odd series $(\Delta_{Sn}+\Delta_{Pb})/(2n+1)$ as well. Therefore it would be revealing to look at two superconductors with almost the same energy gap. Sn and In were the obvious choices. We tried this, but found as Rowell and Feldmann did

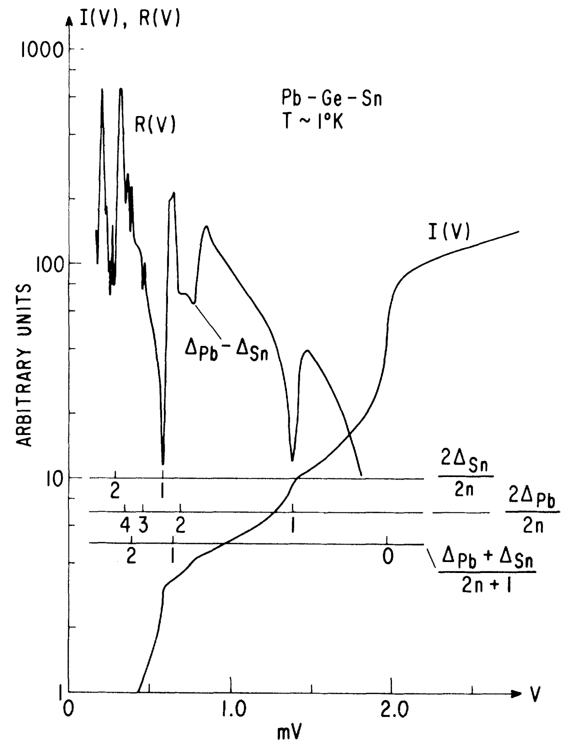

FIG. 12. Current-voltage and dynamic resistance-voltage characteristics of a Pb-Ge-Sn junction. The order of the subharmonic structure which is expected to appear is marked by numbers on the figure. The assignment of the dips to the three different series is numerically difficult because of some overlap, particularly at lower voltages. For example, $\frac{1}{3}(\Delta_{Pb}+{}_{Sn})$ is not apparent because it is too close to $\frac{1}{2}2\Delta_{Sn}$.

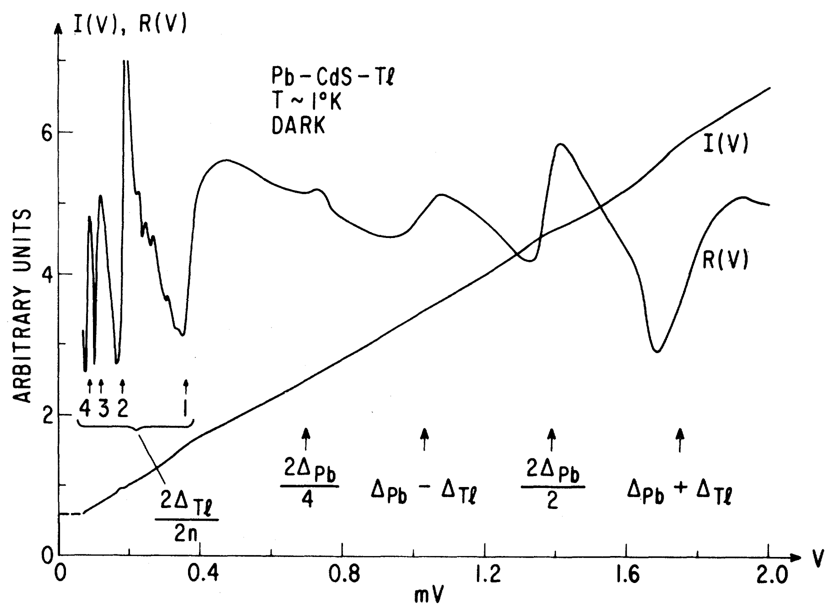

FIG. 13. Current-voltage and dynamic resistance characteristics of a Pb-CdS-Tl junction. The even series is easy to identify, while the odd series is missing.

previously, that the energy gaps were too close together and therefore the current-voltage characteristic looked very similar to an Sn-Sn sample. In some of the samples, however, we could clearly identify a double structure at mid-gap, indicating it to have been caused by $\frac{1}{2}2\Delta_{Sn}$ and $\frac{1}{2}2\Delta_{In}$ rather than by $\frac{1}{2}(\Delta_{Sn}+\Delta_{In})$ which would have given only one structure. By using junctions made of Pb and a Pb-In alloy we obtained a much better resolution, and we can clearly see in these junctions that each gap separately gives an even series while the sum of the two gaps gives an odd series. This result is shown in Fig. 14.

D. Summary of Experimental Results

In this paper we have essentially three new experimental results.

(1) For a strongly coupled junction, the subharmonic structure is proportional to the matrix element squared M^2. It is easy to verify this relationship for the mid-gap bump only, but it is also possible to fabricate junctions where the whole subharmonic series depends linearly on M^2. For weakly coupled junctions the structure increases faster than M^2, but no definite relationship has been established.

(2) The subharmonic structure is very sensitive to externally applied microwave radiation. Typically, the structure is smeared out by the radiation; however, sometimes the radiation appears to split the harmonic structure.

(3) Even though the experimental results get rather complicated, we assert that for two dissimilar superconductors a and b, only the following three series are observed: $2\Delta_a/2n$, $2\Delta_b/2n$, and $(\Delta_a+\Delta_b)/(2n+1)$, where n is an integer.

Again we emphasize that a large amount of data has been collected and essentially discarded for various reasons; however, none of the discarded data contradicts the above summary.

SIMPLE MODEL

We feel that all our results are consistent with the statement that the subharmonic structure is caused by a self-detection of the Josephson radiation. Let us review the simple theoretical argument which leads to this result. Our aim is to demonstrate that it is energetically possible for the Josephson radiation to cause the structure, but we shall not discuss any detailed dynamical process. A more complete discussion is found in Werthamer's paper.[7] When a voltage V is applied across a superconducting junction, Josephson radiation is produced

$$\hbar\omega = 2eV . \tag{1}$$

If we have two dissimilar superconductors a and b, this radiation will excite quasiparticles directly when

$$\hbar\omega = 2eV \geq 2\Delta_a , \quad \hbar\omega = 2eV \geq 2\Delta_b . \tag{2}$$

This gives rise to a structure when the voltage is equal to half of either of the energy gaps in the two superconductors, because the radiation loss must be supplied by the battery.

On the other hand, the Josephson radiation can also be absorbed if we split a Cooper pair but leave

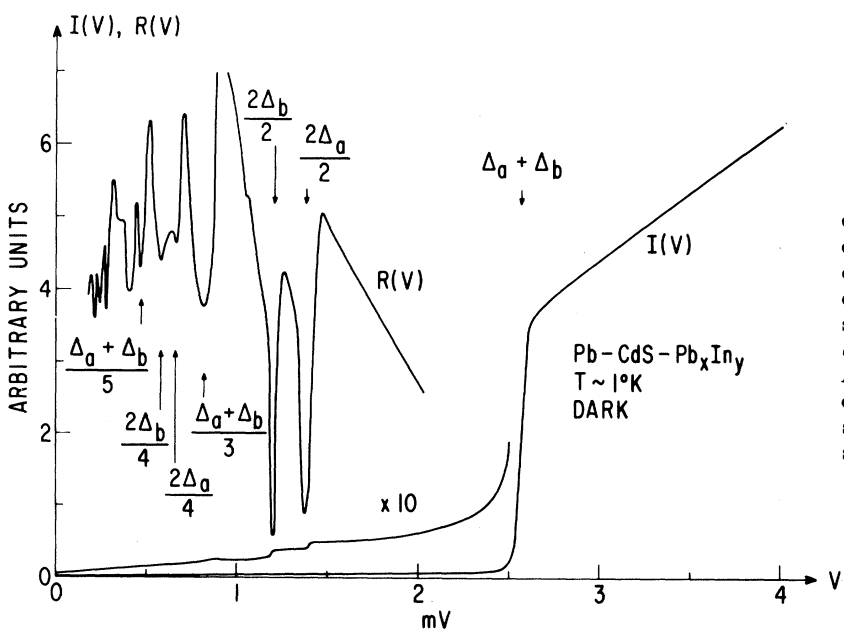

FIG. 14. Current-voltage and dynamic resistance characteristics of a Pb-CdS-$Pb_x In_y$ junction. Because the gaps are approximately equal, the two even series and the single odd series can be identified. a refers to Pb and b to $Pb_x In_y$. Again, at lower voltages it becomes difficult to distinguish between the series themselves and small Fiske steps.

one electron on either side of the barrier in a process similar to the Dayem-Martin mechanism.[20] That process leads to a radiation loss when

$$\hbar\omega = 2eV \geq \Delta_a + \Delta_b - eV \quad (3)$$

and accounts for the structure at

$$eV = \tfrac{1}{3}(\Delta_a + \Delta_b) \,. \quad (4)$$

These results can easily be generalized if we remember that the Josephson equation is nonlinear and equivalent to the pendulum equation. The equation for the phase ϕ is simply given by

$$\ddot{\phi} + (2eI_J/\hbar C)\sin\phi = 0 \,, \quad (5)$$

where I_J is the Josephson current, C is the capacitance of the junction, e is the electron charge, and $\hbar$ is Planck's constant. The voltage across the junction is given by

$$\dot{\phi} = (2e/\hbar)\, V \,. \quad (6)$$

Thus from our knowledge about the pendulum we can schematically plot the voltage as a function of time, as is shown in Fig. 15. For a weakly coupled junction, only the fundamental Josephson frequency will exist, while for a strongly coupled junction, Eq. (1) must be replaced by

$$\hbar\dot{\phi} = 2eV + 2e \textstyle\sum_n V_n \sin(2eV/\hbar)nt \,. \quad (7)$$

Therefore, the photons present in a Josephson junction are given by

$$\hbar\omega = 2neV, \quad n = 1, 2, 3, \ldots \quad (8)$$

which, used with Eqs. (2) and (3), generate for us the complete observed subharmonic structure. Experimentally, of course, it does not matter whether the structure is caused by single photons as indicated by Eq. (8) or by multiple-photon absorption; in fact both processes probably occur.

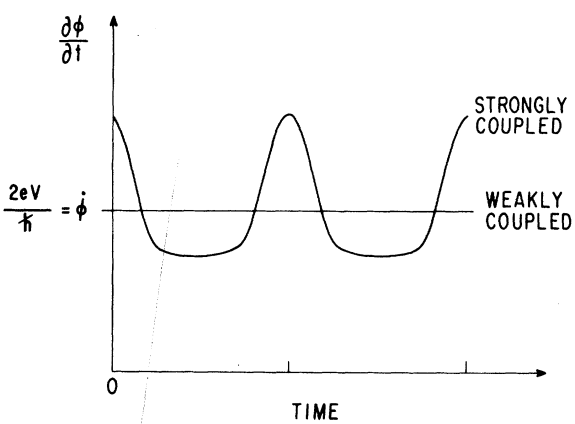

FIG. 15. Schematic illustration of the time derivative of the phase as a function of time. For a weakly coupled system, the Josephson frequency is given by $\hbar\omega = 2eV_A$, while for a strongly coupled system the instantaneous voltage across the junction will be very different from the applied voltage V_A.

DISCUSSION

It frequently has been argued that if the Josephson radiation is responsible for the subharmonic structure, the magnitude should be at most proportional to the fourth power of the matrix element M. At first sight this appears to be reasonable because since the Josephson current is proportional to the square of the matrix element, the radiation density should be proportional to the fourth power. However, this is true only if we linearize the equations governing the problem,[22] i.e., it will be true only for a relatively weakly coupled junction. It is, for example, generally accepted that the current in a Fiske step is due to Josephson radiation, and in strongly coupled junctions this current is clearly proportional to the matrix element squared. We conclude, therefore, that it is improper to linearize the equations governing the system.

Another objection often raised against the simple model outlined is that the even and odd structures appear to have different origins and it is therefore puzzling that the magnitudes and appearances are so similar. To the extent that it is correct to talk about a radiation density in the junction, the absorption coefficients for the two processes causing the even and odd structure have to be of the same order to explain the experiments. It is often argued that the absorption coefficient in the odd case (the Dayem-Martin-like process) should be proportional to the square of the matrix element while the absorption coefficient in the even case is independent of the matrix element and probably of order unity. While this is true in the limiting case of a weakly coupled junction, the absorption coefficient for the odd process approaches unity in the other limit of a strongly coupled junction, resulting in an even and an odd structure of the same order of magnitude. The only way these questions can be properly dealt with is to solve the complete problem outlined by Werthamer.

Our Fig. 4 shows that the mid-gap structure for a weakly coupled junction increases faster than M^2 and then for a stronger-coupled junction becomes proportional to M^2. This is very similar to the known behavior of the Josephson current. For a weakly coupled junction, the Josephson current is unobservable because the binding energy is less than the thermal energy, while for strongly coupled junctions the Josephson current is proportional to M^2. The dc Josephson currents which we observe in our samples are much smaller than the theoretical maximum. We believe the main reason for this to be that the junctions are very nonuniform and therefore the self-field from the current makes several of the ac modes more stable than the dc mode.

Rowell and Feldmann divided the observed structures into two classes based on appearance. In this respect we must remember that no two junction characteristics are exactly alike; thus we should be careful when we talk about "line shapes." However, it is true that in some junctions the structure can be described as cusplike while in others the structure is more like a current step. The mid-gap structure in Fig. 6 is cusplike in the dark state of the junction, steplike in the light-saturated junction. The cusplike behavior is caused by small areas in a junction where the matrix element is, relatively speaking, very large. Because of the strong coupling, the Josephson equation is very nonlinear and higher-order processes ($n > 1$) are readily observable. The ac Josephson current flowing through such an area is much larger than the single-particle tunneling and in some extreme cases dominates the total current-voltage characteristic as illustrated in Fig. 7. Rowell and Feldmann postulated that these kinds of characteristics are due to metallic shorts, but we feel that it is more an intermediate case between a simple tunnel junction and a metallic short. (In fabricating such junctions, we find that in order to avoid a metallic short we always must expose the junction to air before the top metal layer is deposited.) The steplike structure is due to a relatively weakly coupled junction where large areas contribute to the Josephson radiation. The radiation density in a junction is maximum at thin spots and inhomogeneities in the barrier. Even in the case where thin spots do not dominate the current, they may be important for the radiation absorption; since they have a slightly different gap this could cause a smearing of the half-gap and higher-order structure. The structure is never very sharp in these junctions and commonly only the mid-gap structure is readily visible. Another way to understand the appearance of the structure is to assume that the Josephson frequency itself is not very sharply defined. Physically this seems plausible if we view the voltage across the junction to be caused by the motion of flux analogous to the commonly accepted view of a type-II superconductor. In a large junction a steady-state solution of the flux motion is improbable, except in a highly idealized junction. Because of irregularities in the junctions such as an uneven oxide thickness, the flux motion would be irregular and the motion would be activated by thermal fluctuations and other noise sources. The instantaneous voltage will vary over the area of such a junction causing a variation in the frequency. This inherent smearing will render higher-order structure difficult to observe. We emphasize that this effect is quite different from the frequency modulation which is obtained in a self-consistent treatment of the Josephson effect; the latter is, of course, present in all junctions. Thus in spite of the two different appearances of the structure, we will argue that in both cases the Josephson effect is the direct cause.

This conclusion is supported by the fact that externally applied microwave radiation has a pronounced effect on the structure. In our simple model we should now get additional structure, for example, when the sum of the external radiation $\hbar\omega$ and the internal radiation $2eV$ equals the energy gap 2Δ. If higher-order processes are taken into account we obtain $2neV + m\hbar\omega = 2\Delta$, where n and m are integers. Thus the simplest process is at least of second order. The results in Fig. 10 can be accounted for by using $n = 1$ and $m = 2$ in the above relation. Why the simpler process, where $m = 1$, does not dominate is somewhat surprising; however, it may be explained several ways, for instance, by the geometry of the junction. We should remember that the junction is not uniform and that the effective area may be located at a spot where the $m = 2$ modes couple much more effectively. The typical result is simply a smearing of the structure which is consistent with a nonuniform coupling of the external to the internal radiation.

We want to emphasize that we are talking only about the energy-conserving part of the process, and we are not prepared to discuss the dynamics of the process in any detail. Energetically it does not matter whether the tunneling pairs create electromagnetic radiation which then in turn is absorbed, or whether the tunneling pairs somehow convert their energy directly into excitations in the superconductors. Also, it does not matter whether the external radiation interacts directly with the pair current, or whether the unperturbed Josephson radiation combines with the external radiation as considered above. We also realize that it may be awkward to speak of a two-photon absorption process as we did when the externally and internally generated radiation is combined to create excitations, but we are at present unable to deal with the problem in greater detail. Werthamer has done this in his paper; however, as far as we know, the equations have never been numerically solved.

In the third experiment we carried out we were able to confirm Werthamer's prediction that for two unequal superconductors we should obtain two even series and one odd series. This is a natural result if the Josephson effect is the cause of the structure as shown more or less schematically in our model.

Finally, the explanation is consistent with the fact that the structure is relatively insensitive to applied magnetic fields. Only the total dc Josephson current is strongly dependent upon the applied magnetic field; the maximum amplitude of the current density is not magnetic field sensitive. It is somewhat surprising that the structure can appear in junctions which display little or no dc Josephson current. We have verified experimentally that when the current mainly flows through thin spots in a junction, the total dc Josephson current is more magnetic field sensitive than for a uniform junction. One reason for this, we believe, is that high local self-fields make the ac modes more stable at a fixed dc current through the sample. Thus, the nonexistence of a dc Josephson current does not imply that there is no pair tunneling.

CONCLUSIONS

In this paper we have attempted to show that the subharmonic structure often observable in a tunneling experiment between two superconductors is due to the Josephson effect. We have three new experimental facts which support this conclusion: (1) For strongly coupled junctions, the structure is proportional to the tunneling matrix element squared. (2) The observed subharmonic structure is very sensitive to externally generated microwave radiation. (3) There is a distinction between structure at $2\Delta/n$ depending on whether n is even or odd.

We find it somewhat puzzling that the odd and even subharmonic series have the same magnitude; however, we expect that this is contained in the nonlinear equations describing the problem.

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[1] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).
[2] I. Giaever, H. R. Hart, Jr., and K. Megerle, Phys. Rev. 126, 941 (1962).
[3] B. N. Taylor and E. Burstein, Phys. Rev. Letters 10, 14 (1963).
[4] J. R. Schrieffer and J. W. Wilkins, Phys. Rev. Letters 10, 17 (1963).
[5] I. K. Yanson, V. H. Svistunov, and I. M. Dmitrenko, Zh. Eksperim. i Teor. Fiz. 48, 976 (1965); 47, 2091 (1964) [Soviet Phys. JETP 21, 650 (1965); 20, 1404 (1965)].
[6] J. M. Rowell and W. L. Feldmann, Phys. Rev. 172, 393 (1968).
[7] N. R. Werthamer, Phys. Rev. 147, 255 (1966).
[8] B. D. Josephson, Advan. Phys. 14, 419 (1965).
[9] S. M. Marcus, Phys. Letters 19, 623 (1966); 20, 236 (1966).
[10] J. W. Wilkins, in *Tunneling Phenomena in Solids*, edited by E. Burstein and S. Lundquist (Plenum, New York, 1969).
[11] I. Giaever, Phys. Rev. Letters 20, 1286 (1968).
[12] I. Giaever and H. R. Zeller, J. Vacuum Sci. Technol. (to be published).
[13] D. M. Warschauer and C. D. Reynolds, J. Phys. Chem. Solids 23, 457 (1960).
[14] M. R. Lorentz, B. Segall, and H. H. Woodbury, Phys. Rev. 134, A751 (1964).
[15] H. C. Wright, R. J. Downey, and J. R. Canning, Brit. J. Appl. Phys. 1, 1593 (1968).
[16] I. Giaever, in Ref. 10.
[17] G. I. Rochlin, Phys. Rev. 153, 513 (1967).
[18] I. Giaever, Phys. Rev. Letters 14, 904 (1965).
[19] S. Shapiro, Phys. Rev. Letters 11, 80 (1963).
[20] A. Dayem and R. J. Martin, Phys. Rev. Letters 8, 246 (1962).
[21] M. D. Fiske, Rev. Mod. Phys. 36, 221 (1964).
[22] R. E. Eck, D. J. Scalapino, and B. N. Taylor, Phys. Rev. Letters 13, 15 (1963).