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Superconducting Thin-Film Detector of Nuclear Particles*

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Superconducting films of tin and indium on supporting substrates have functioned successfully as detectors of individual nuclear particles. The films are sufficiently thin and narrow that individual α-particle impacts initiate superconducting to normal transitions which spread, in the presence of a transport current, to span a full film cross section. The transitions are observed by means of the ir drop produced by a transport current. For low current densities self-terminating voltage pulses of a few nsec duration are observed. At higher current densities the boundaries of a normal region initiated by an α particle propagate by Joule heating to the ends of the film. The range of the 5.3-MeV α particles utilized for these experiments greatly exceeds the 0.1 μm thickness of the films and the resulting deposition of energy in the substrate affects the response of any film which is in direct contact with its substrate. The introduction of a thin thermally insulating layer of varnish between the film and its substrate, which in these experiments was either glass or crystalline quartz, increases the thermal decay constant to the extent that the film is thermally isolated from its substrate for a period comparable to that of pulse formation. The variation of count rate with film current has been studied and is shown to be consistent with the variation of critical current density along the length of the film. This model, coupled with classical heat diffusion within the film, accounts for the observed behavior of the thermally isolated films.

I. INTRODUCTION

This paper reports the results of experiments on some of the transient effects of individual nuclear-particle impacts on superconducting films. In particular, superconducting films of tin and of indium have been bombarded with 5.3-MeV α particles and the properties of the normal regions initiated by the individual α particles have been determined. The films are sufficiently thin and narrow that the energy deposited by an individual α particle can initiate a superconducting-to-normal transition across a full film cross section. The transitions are observed by means of the ir drop produced by a transport current. The primary normal region, produced by the energy lost by the α particle, need not cross a complete section of the film in order for its effect to be observed because any remaining width of the film will go normal provided the transport current is such that the critical current density is exceeded. Figure 1 illustrates the current flow schematically. The size of the normal regions is conveniently expressed in terms of an effective radius, defined as the radius of a hypothetical region which, if physically removed, would result in the same current-capacity reduction as does an α-particle traversal. So defined, the effective radius is somewhat larger than the radius of the normal region because of the temperature increase in the adjacent material.

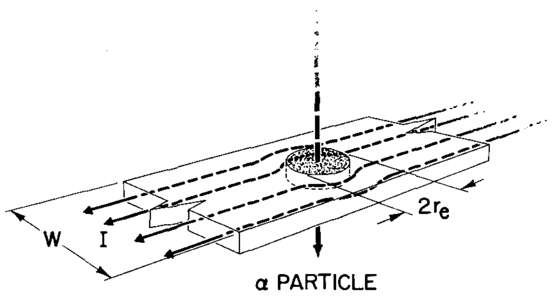

FIG. 1. Schematic representation of the presumed current paths following an α-particle impact.

Some earlier results were published elsewhere.[1] The experiments are similar to those proposed by Sherman[2,3] and to the work of Andrews, Fowler, and Williams[4] who obtained some evidence of effects of individual particles, using much wider films.

A variety of rectangular films ranging in width from 5 to 50 μ and in thickness from 500 to 2000 Å were evaporated onto either glass or crystalline quartz substrates. For sufficiently low current densities the normal regions initiated by α particles return spontaneously to the superconducting state. The duration of these self-terminating or self-recovering voltage pulses is of the order of tens of nsec. At higher current densities the normal regions do not return automatically to the superconducting state, but instead expand, because of Joule heating, to encompass the whole film.

The film thicknesses employed are less than the range of the α particles so that energy is deposited in the substrate as well as in the film. In the case of the crystalline quartz substrate, where the phonon mean free path is of the order of 1 cm, the evidence indicates that ballistic phonons originating in the quartz carry enough energy back into the detector to significantly increase the radii of the regions driven normal. For glass substrates, where the phonon mean free paths are short and energy transport is consequently diffusive, the evidence shows again that heat exchange is significant. Before the detector-substrate heat exchange was recognized as a factor the early results,[1] all for quartz substrates, were puzzling in that the effective radii of the normal regions were too large to be consistent with the results of a heat-flow calculation for an isolated film. By coincidence the observed effective radii agreed reasonably well with radii calculated on the basis that only the electron specific heat of the metal film was involved, with the lattice heat omitted. The calculated characteristic time of such a heat pulse was of the order of 10^{-10} sec, which was not small enough to make the assumption of lack of equilibrium between the electrons and lattice reasonable. This made it seem likely that heat exchange with the substrate was responsible for the anomalous behavior.

To examine the extent of substrate involvement, experiments were performed in which pairs of very closely spaced parallel films were evaporated onto glass and quartz substrates. The films were electrically independent and thermally connected only through the substrate. If, as was expected, the substrate heat was reaching the film, then for such closely spaced narrow films one would expect to observe coincident pulses from the two films. When these experiments were carried out a significant fraction of the events observed were coincident; as a consequence, it was evident that the assumption of total film-substrate independence was invalid.

In an effort to thermally isolate the metal film, a thin varnish layer was introduced between the films and their substrates. Calculations had suggested that the introduction of this acoustic impedance mismatching layer could reduce the transmission of incident phonon energy by more than an order of magnitude. Indeed, with these layers, the rate of coincidences for the pairs of films was reduced essentially to zero, and the effective radii approached the values expected for heat flow within the film alone.

Computer solutions of the heat-diffusion equation for the metal films, under the assumption of no heat exchange with the substrate, give values of the effective radii of the normal regions in good agreement with the measured values for both indium and tin on varnish-coated quartz and glass substrates. The films of both tin and indium are made with a high impurity concentration to provide a relatively short electron-transport mean free path of about 0.1 μ at 4 K. The observed effective radius of the region heated above the superconducting critical temperature is of the order of 5 μ. Thus the mean free paths involved are small enough to make the classical heat-diffusion calculations reasonable for the heat flow within the film.

II. EXPERIMENTAL

The detectors were manufactured by evaporation of the film material through razor-blade masks onto glass or crystalline quartz substrates. The substrates were 0.32-cm-thick by 2.54-cm-diam disks with the parallel surfaces optically polished. In the case of the crystalline quartz, the faces of the disks were perpendicular to the c axis. The evaporation was carried out at a rate of about 20 Å/sec in a vacuum of the order of 10^{-6} Torr. These pressures and slow deposition rates result in film conductivities of around 10^6 mho/cm at 4 K. Such relatively low conductivities are desirable from the standpoint of detection sensitivity and also provide the short electron mean free path necessary to make the classical heat-flow calculations tenable. The films were evaporated onto substrates held at 0 °C for tin and at −80 °C for indium. The thickness of the films was monitored during deposition by a quartz-crystal oscillator.

In the experiments which required pairs of closely spaced parallel films, the films were evaporated simultaneously through a razor-blade mask. The gap between films was created by masking the center with a fiber drawn from Duco cement.

Varnish films were made by the electron bombardment technique described by Christy.[5] In this method the varnish film is formed in vacuum through the polymerization, by electron bombardment of backstreaming diffusion-pump oil which impinges on the substrate surface when the oil vapor is permitted to enter the system by warming the trap. Varnish film thicknesses, which in these experiments were always 300 Å, were determined from bombardment time, at constant current, after interferometric calibration. The pump oil was Dow-Corning 704.

During measurement of the α-particle pulses the specimen was cooled by a liquid-helium bath on the backside of the substrate. Temperatures were determined by measuring the vapor pressure of the bath. A thin Po-210 source, suspended in vacuum above the film, supplied α particles. Generalized heating of the film and substrate was avoided by keeping the α-particle flux low.

Electrical contact with the sample was made through miniature 50-Ω coaxial cables which were terminated, when required, in a 50-Ω resistor consisting of fine nichrome wire which replaced the final 1.5 cm of the central conductor. Low-temperature resistances and transition temperatures were measured by the usual four-terminal technique in which a 50-μA current was employed.

The self-terminating pulses are typically a few hundred μV in amplitude and have rise times of the order of 10 nsec except for films on bare glass substrates where the rise times are sometimes as long as several hundred nsec. Pulse decay time is strongly current dependent, increasing steadily with current until Joule heating is sufficient to cause the boundaries of the initial re-

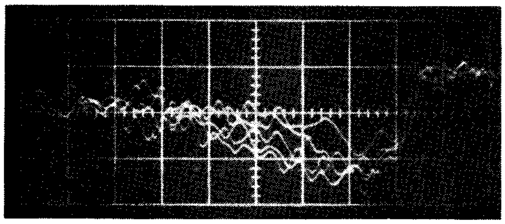

FIG. 2. Self-recovering voltage pulses for a tin film on a crystalline quartz substrate. Vertical scale is 100μV/large division and the horizontal rate is 20 nsec/large division.

sistive region to propagate to the ends of the film. Observation of the self-terminating pulses is accomplished by using a Keithley 107 pulse amplifier as a preamplifier driving a Tektronix 585 oscilloscope. The rise time of this combination is less than 5 nsec and has a noise equivalent referred to the input of about 30-μV rms. Traces of a number of self-recovering pulses are shown in Fig. 2.

To record pulse rates for transport currents sufficient to cause propagation of the boundaries of the normal region, it is necessary to momentarily interrupt the film current after each pulse in order to permit recovery of the detector to the superconducting state. Switching is sufficiently rapid that current cutoff occurs within 0.1 μsec of the time the film voltage exceeds a predetermined level. The current is turned back on after a dead time determined by the requirement that the pulse rate, corrected for this dead time, be independent of the dead time. For the pulse rates used in these experiments, 300/min maximum, dead times of the order of 1 msec fulfill the requirement. A dead time of 10 msec was employed, however, to absolutely insure that the heat generated by these pulses did not affect the results.

The critical currents of the detectors in the absence of α-particle bombardment were measured using a combination of dc and pulsed current techniques. We observe, as have others[6] that the critical current of a rectangularly shaped superconducting film appears to vary along its length. The so-called dc critical current, measured by slowly increasing film current until the first resistance appears, characterizes only the "worst" segment of the film, while other segments have a higher critical value. This behavior seems likely to be associated with establishment of an intermediate-state domain pattern within the film, but the simpler model of variation of critical current with location along the film is capable of providing an approximate prediction of the count-rate—vs—current data. It was not possible to determine a functional relationship between critical current and position along the length of a film in the sense that to each position a critical current could be assigned. It was possible, however, to estimate the fraction of the film length that had had its critical current exceeded as a function of current. To do this this, a series of fast-rising current pulses was applied to the film, and the time variation of voltage observed. Film current was monitored with a current transformer driving one beam of a dual-beam oscilloscope. Voltage was displayed on the other beam, and the two traces were photographed together in families in which the current was increased in steps. One such photograph is shown in Fig. 3 where the currents are displayed on the upper beam and the film voltages on the lower. The current pulses applied to the film rose in about 1 nsec. This does not show on the current monitoring traces which were displayed at the relatively slow rate of 20 nsec per large division after the pulses had passed through an amplifier with a rise time of 13 nsec. The sweep rate for the voltage pulses was 2 nsec per large division. The rise time of the voltage pulses displayed was limited to about 2 nsec by an amplifier. For the lowest current pulse, very little of the film has switched within 15 nsec of the arrival of the current pulse, which time is conveniently indicated by the inductive pulse preceding the rise in film resistance. As the current is increased, an increasing fraction of the film switches more and more rapidly until, as indicated in the top trace, essentially the whole film is switching at a rate equal to or greater than the observational rate limitation imposed by the equipment. Just how much of the film has had its critical current exceeded at any given current can be, at best, only estimated from these data. At currents in excess of dc critical, the film switches at multiple points along its length and each normal segment promptly begins expanding because of Joule heating. At the current levels of interest the expansion rate is sufficiently rapid that it is difficult to determine what fraction of a film has switched because its critical current has been exceeded and what fraction has switched because it is immedi-

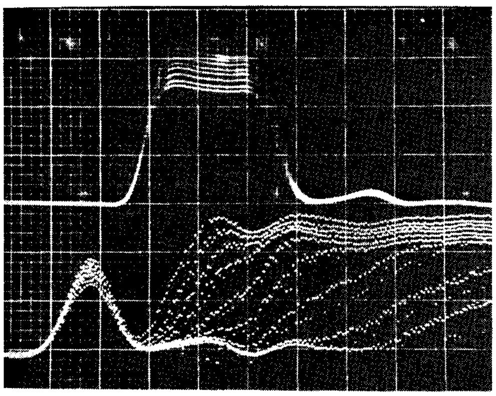

FIG. 3. Switching characteristics of a quartz-backed tin film. Upper beam: film current vs time. 10 mV/division and 20 nsec/division. Lower beam: film voltage as a function of time. Vertical: 10 mV/division; horizontal: 2 nsec/division. The initial pulse on the lower beam is the inductive voltage from current rise in film.

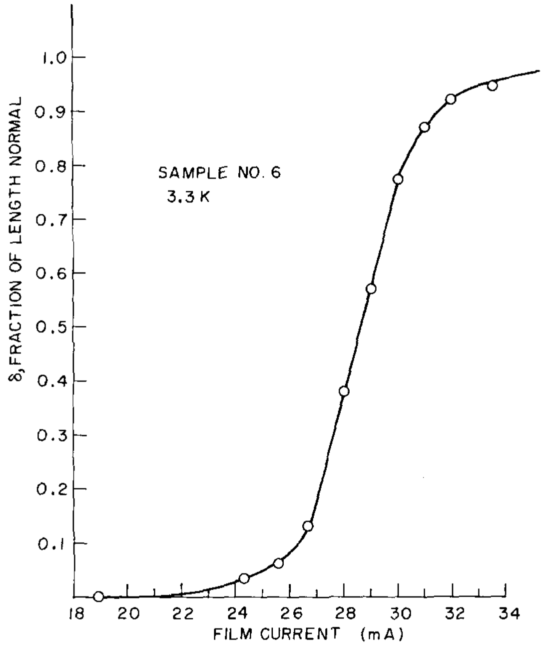

FIG. 4. Fraction of the film length which enters the normal state (within 7 nsec of current-pulse arrival) as a function of pulse-current magnitude. The data shown are taken from the photograph of Fig. 3.

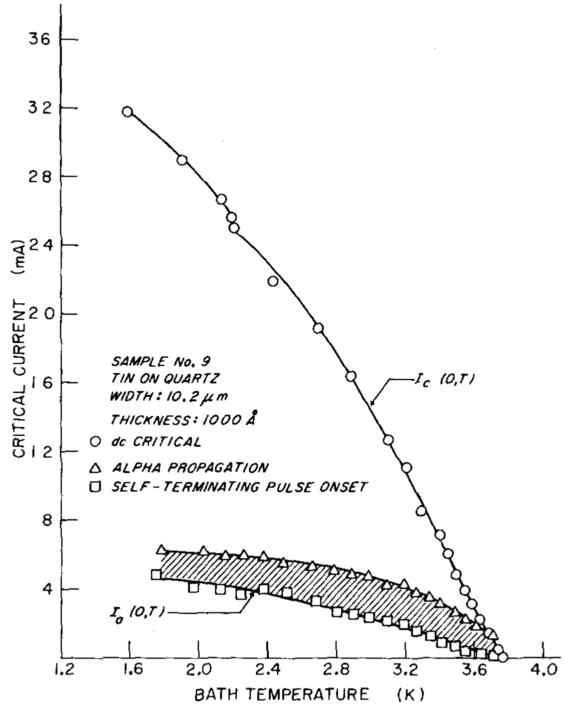

FIG. 5. Critical currents of a 10.2-μm-wide tin film on a crystalline quartz substrate. There was no interposed varnish layer on this sample. The discontinuity in I_c occurred at the helium λ point.

ately adjacent to a switched region from which it is receiving heat. In spite of this difficulty it was found that if the critical current was taken as that which caused a given fraction of the film to switch normal within 7 nsec of the time the current pulse reached its maximum value, then the count-rate dependence on film current could be adequately described. The critical current so derived is written $I_c(\delta, T)$, where δ represents the fraction of the film length which has had its critical current exceeded at the current $I_c(\delta, T)$, while at a bath temperature T. Then $I_c(0, T)$ is the minimum or dc critical current and $I_c(1, T)$ is the maximum critical current. Values of δ derived from the curves of Fig. 3 are plotted as a function of $I_c(\delta, T)$ in Fig. 4.

III. RESULTS

The general regions of α-particle counting behavior for two different film widths of tin are indicated in Figs. 5 and 6. The behavior of indium films is similar except that the critical temperature is at 3.4 K. The solid curves show $I_c(0, T)$ while the dashed curves labeled $I_a(0, T)$ are the minimum currents at which the effect of α particles is observed. Self-terminating pulses occur only in the shaded regions shown. The lower boundaries of these regions define the currents $I_a(0, T)$ at which α-particle-induced voltage pulses first appear. Propagation occurs at the upper boundaries.

In Fig. 5, where the film width is small, self-terminating pulses occur over the whole range of temperatures examined. As the film width is increased, however, the region of the self-terminating pulses narrows, as shown in Fig. 6, where, at a temperature of 3.4 K and below, the pulses propagate at onset. From these results it seems reasonable to consider the effect of an α-particle traversal as

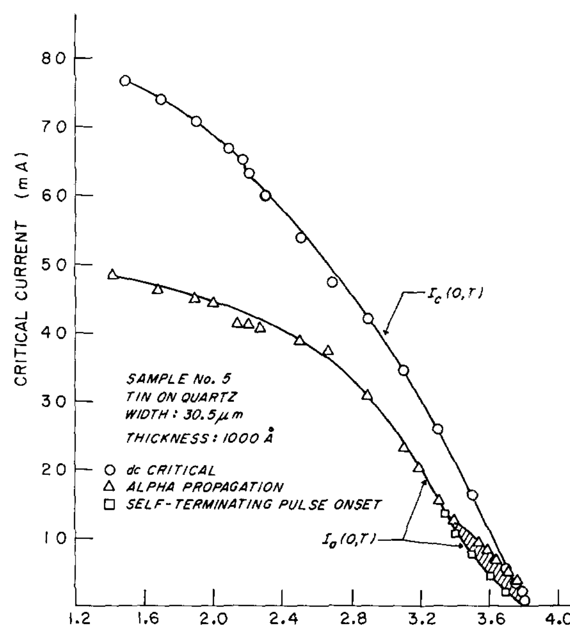

FIG. 6. Critical currents of a 30.5-μ-wide tin film. The substrate was unvarnished crystalline quartz. The discontinuity in I_c occurred at the helium λ point.

equivalent to a transient reduction of the current-carrying width of the film at the point of α-particle impact.

The effective radii of the primary normal regions produced by the α particles can be obtained from data such as depicted in Figs. 5 and 6, if certain simplifying assumptions are made. The critical current for a thin film varies approximately linearly with width[7] even though the current density is peaked at the edges.[8] Since the details of current flow around a primary normal region produced by an α particle have not been determined analytically, the simplifying assumption is made that the impact of an α particle reduces the superconducting width of the film by twice the effective radius of the normal region. On this basis, if $I_a(0, T)$ is the temperature-dependent minimum transport current required to produce a resistive pulse when an α particle traverses the film, then the effective radius $r_e(T)$ of the region driven normal is given by the relation

$$I_a(0, T) = I_c(0, T)[w - 2r_e(T)]/w, \tag{1}$$

where w is the geometric film width. In other words $I_a(0, T)$ is interpreted as the critical current, at a bath temperature T, of a film whose width has been reduced by an amount $2r_e(T)$.

Values of $r_e(T)$ can be obtained by use of Eq. (1). However, more consistent values are obtained by an extension of this method which does not involve the necessity for the direct observation of the threshold current $I_a(0, T)$. Such a threshold is difficult to obtain because of the low counting rate and small slope

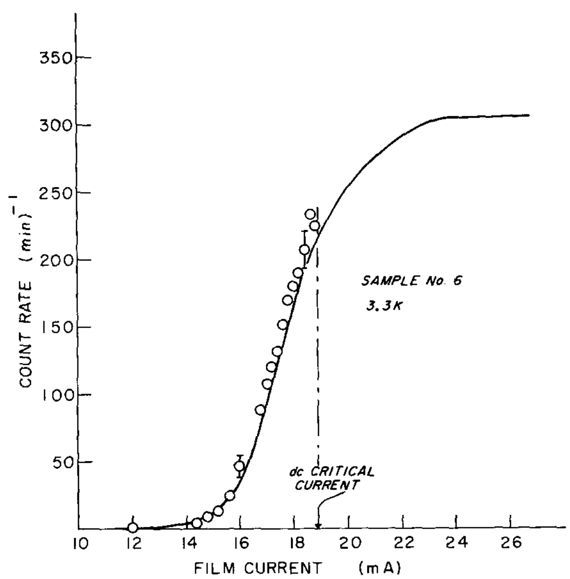

FIG. 7. Count rate as a function of current at 3.3 K. The circles are experimental points and the solid curve is theoretical. At this temperature the dc critical current of the sample was 18.8 mA so that the count rate above this level could not be measured. The error bars are the one-sigma counting statistics.

at the point of intercept with the axis. The extension involves the manner in which the pulse rate varies with current.

As the transport current is increased from zero, pulses first appear at a current of $I_a(0, T)$. The pulse rate increases reversibly with increasing transport current above this threshold, as shown in Fig. 7. This behavior suggests that the critical current in the absence of α particles is a function of position along the film. Making this assumption, and using data such as that shown in Fig. 4 for δ, a relationship between film current and the fraction of the film sensitive to α particles can be obtained by numerical integration. From this information the variation of count rate as a function of current can be predicted for a given source strength. A first-order correction for the effect of impact near the film edges is readily included. The solid curve shown in Fig. 7 is the result of such a calculation.[9] There usually was an upper limit to the range of currents over which the count rate could be measured, imposed by the dc critical current. Thus it was not generally possible, as illustrated in Fig. 7, to measure the count-rate curve out to full saturation.

The preceding model and the pulse critical-current data, as displayed in Fig. 3, permit determining associated values of $I_a(\delta, T)$ and $I_c(\delta, T)$ for values of δ above the initial region of very small slope. For these pairs of values, a modification of Eq. (1) applies:

$$I_a(\delta, T) = I_c(\delta, T)[w - 2r_e(T)]/w$$

or (2)

$$r_e(T) = \tfrac{1}{2}w[1 - I_a(\delta, T)/I_c(\delta, T)].$$

This permits determination of $r_e(T)$ in a more convenient and dependable pulse-rate range. Values of $r_e(T)$ were determined by this means for points below the middle of the pulse-rate curve and are consistent with values obtained at the counting threshold using Eq. (1).

An independent method of determining the effective radius is available through utilization of the pulse-height voltage. For transport currents substantially below the current for which continuous boundary propagation commences, the normal region initiated by an α particle would be expected to expand until it has spread to the full width of the film and has a length approximately equal to the diameter of the circle of effective radius r_e. Under this assumption, the pulse height voltage V should be

$$V = 2r_e I/\sigma A,$$

where σ is the film conductivity and A is the cross-sectional area of the film. Experimentally, the pulse heights are found to be proportional to I, up to values of I just below those that cause continuous

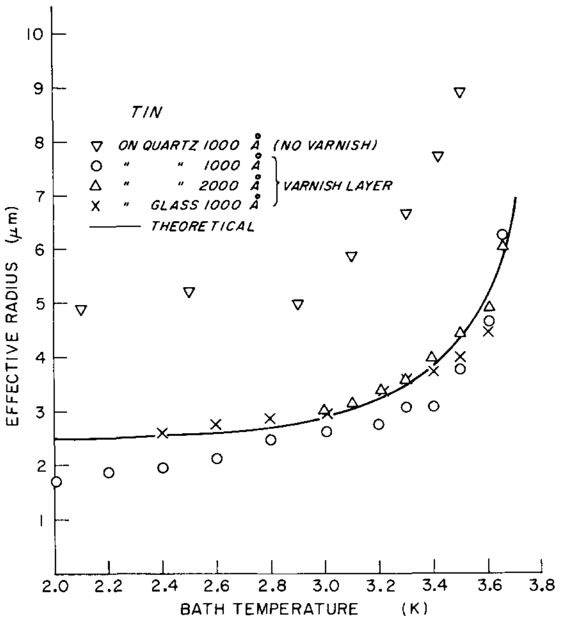

FIG. 8. Experimental and theoretical effective radii in tin as a function of temperature.

boundary propagation. Values of r_e can be obtained in this way for temperatures closer to the zero-current critical temperature than the method of obtaining r_e from the critical values.

Values of effective radii determined by these two independent methods are shown in Fig. 8 for tin films on both bare and varnish-coated quartz substrates. The behavior for indium films is similar except that the effective radii are smaller because of the larger heat capacity of indium. The solid curve in Fig. 8 represents the effective radii predicted by a computer solution of the heat-diffusion equation, under the assumption of no heat interchange with the substrate. The temperature dependences of both the electron and lattice specific heat and the thermal conductivity were taken into account for both the superconducting and normal regions. The experimental points lie close to the theoretical curve for the varnish-coated substrates. This indicates that the varnish layer has succeeded in thermally isolating the film for a time comparable to the pulse rise time (about 10 nsec). The points for the bare quartz substrate lie far above the theoretical curve. This indicates that there has been a net heat flow from the substrate to the metal film across the interface. This presumably takes place because the range of the incident α particles extends deeply (about 10 μm) into the quartz, and the relaxation of the initial electron excitation in the quartz gives rise to a large flux of phonons. The long mean free path of these phonons at the temperature of about 3 K provides considerable heat transport to the film during a time interval comparable to the time for radial heat flow within the metal film.

The theoretical curve in Fig. 8 was calculated with the assumption that the energy had been deposited in a small cylindrical volume surrounding the path of the α particle. The results are independent of this initial temperature distribution. More detailed results of these calculations are illustrated in Fig. 9 for tin at a bath temperature of 3.4 K. Curve A represents the radius of the region for which the temperature has been raised to the critical temperature, plotted as a function of time. This radius is not the effective radius r_e, however, because no account has been taken of the material adjacent to the normal core which, although remaining in the superconducting state, has been heated and therefore has a reduced critical-current density. If the critical-current dependence on temperature of the material surrounding the slumping thermal spike is assumed to vary in the same manner as for a complete film, then the added effect due to the heated surroundings can be calculated. The results of such a calculation are shown by curve B of Fig. 9 which shows the time dependence of the equivalent added radius due to critical-current reduction in the adjacent material. The sum of these two curves is shown as curve C. The maximum point of this curve is the predicted effective radius for that particular bath temperature. The solid curve in Fig. 8 gives the values of r_e calculated in this manner.

Films on bare glass behave differently from films on bare quartz, or varnished quartz, or glass. Individual pulse rise times for glass vary from 20 to 300 nsec, with all other conditions constant. This appears to be the result of heat transfer from the glass to the film and rather slow heat diffusion radially in the glass. The faster-rising pulses correspond to α-particle impacts directly on the metal film, whereas the slower-rising pulses are the result of off-film, or near miss, impacts. Experiments with pairs of parallel films also confirm this view. The relative rise times and time delays between "coincident" pulses are consistent with calcu-

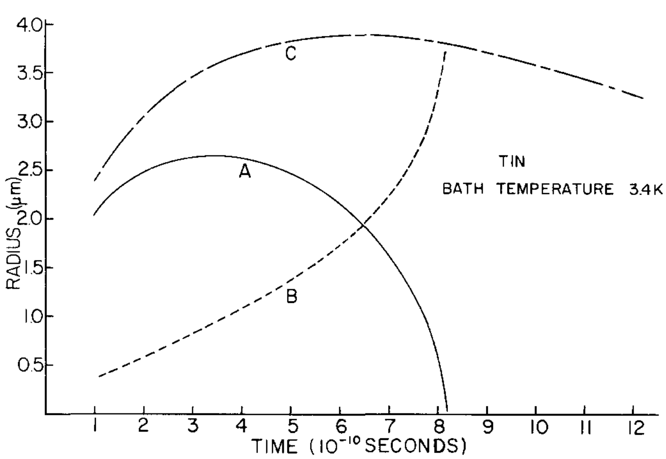

FIG. 9. An example of the results of computer calculation of the effective radius in tin. The calculation predicts a maximum effective radius of 3.9 μm at 6.6×10^{-10} sec.

lated times for heat diffusion in the glass. Ordinary heat diffusion is applicable in the glass because the phonon mean free path is much smaller than the dimensions involved.

Interposing a varnish layer between a glass substrate and a pair of parallel films reduces the number of coincidence pulses in the pair nearly to zero. This indicates that the varnish layer has reduced the heat exchange with the substrate, thermally isolating, at least on a short time scale, the metal film from the glass substrate. The recovery is not as fast for varnish on glass as for varnish on quartz because the glass substrate is locally hot by heat conduction of the α-particle energy within the substrate. By contrast, in the case of quartz the α-particle energy in the substrate is dispersed over a wide region at the speed of sound by the ballistic flight of very long mean free path phonons. Thus for a practical counter, a quartz substrate or other long mean free path material would be preferable to glass because of the more rapid recovery.

As a further check on the thermal isolation afforded by the varnish layer, films of a variety of thicknesses were prepared on varnish-coated quartz. Results for two thicknesses are shown in Fig. 8. The observed effective radii are independent of film thickness. This is what would be expected for thermal isolation as the heat is deposited by the α particle at a uniform rate per unit length of track near the beginning of the track, and the heat flow within the film is essentially radial.

IV. SUMMARY

These experiments demonstrate that superconducting thin films can be used to detect α particles. Both self-recovering and propagating pulses are observed. The count-rate behavior as a function of film current is expressible in terms of a variation of of critical current along the length of the film, and a variation of sensitivity as a function of lateral position of the α particle's impact on the film. Pairs Pairs of adjacent films deposited on bare quartz show coincident pulses due to heat transfer from the substrate. If a varnish layer is interposed, the coincidence rate is reduced nearly to zero. For superconducting films on bare quartz the effective radii are larger than for films on varnish. This indicates that phonons, generated by relaxation of electron excitation in the quartz, transfer more heat to the film than the film loses to the quartz. The varnish films provide high reflectivity for phonons because of the poor acoustic impedance match at the quartz-varnish and varnish-metal interface. For device purposes the bare quartz substrate provides better sensitivity (larger area and larger pulse heights) than a varnished quartz substrate, although prediction of the expected radii of the normal regions is more empirical. For superconducting films on a varnish layer, for which the heat exchange with the substrate is greatly reduced, the effective radii of the normal regions are consistent with values predicted on the basis of heat flow in cylindrical symmetry in the film, incorporating the temperature variation of heat conductivity and specific heat in both the superconducting and normal regions. Further evidence of the effectiveness of the short-term thermal isolation provided by the varnish is that the effective radii of the normal regions are independent of film thickness for films on varnish.

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