A STUDY OF SUPERCONDUCTING TIN BY THE TUNNEL EFFECT

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Submitted to JETP editor June 19, 1963

J. Exptl. Theoret. Phys. (U.S.S.R.) 45, 1839-1849 (December, 1963)

The width of the gap in the electron energy spectrum of tin in the superconducting state is studied by the tunnel effect method. Depending on the crystallographic orientation, the gap width changes from $2\Delta/kT_c = 2.7$ to $2\Delta/kT_c = 4.3$. The anisotropy of Δ is of a complicated type. Together with large regions (~ 15 angular degrees) in which the change of Δ does not exceed 2%, regions are observed in which there is a sharp change of Δ, amounting to 20% in intervals of $\sim 5°$. It is suggested that anisotropy of such a type is related to the structure of the Fermi bands of tin. The relative change of the gap width with temperature is close to that implied by the theory of superconductivity.

THE properties of tin in the superconducting state were studied in this work by the tunnel effect method. In distinction from previous work (e.g. [1-3]) the subjects of the study were single-crystal specimens of such high purity that the mean free path of electrons exceeded by a large factor the characteristic dimension of pairs in the superconductor. This allowed the observation of new effects associated with anisotropy in the properties of tin. (Preliminary results were published in [4].)

METHOD

The properties of a superconductor S can be determined from the tunnel effect both between a superconductor and a normal metal (S — n) and between two superconductors (S — S*). The density of states distribution for electrons close to the limiting energy is given in Fig. 1 for these two cases.[1)]

It is apparent that at $T = 0$ there is no tunnel current for the system S — n up to the voltage $eV = \Delta$, and for the system S — S* to $eV = \Delta + \Delta^*$ (Δ is the width of the gap in the electron energy spectrum of the superconductor). For the S — n system the sharpness of the increase in current at $eV = \Delta$ is determined by the distribution of electrons close to the limiting energy in the normal metal, and sufficiently sharp results can only be obtained in the region of limitingly low temperatures where $\Delta \gg kT$. For example, in tin an uncertainty of $\sim 1\%$ occurs already at several tenths of a degree Kelvin. For the S – S* system the sharp increase of current at $eV = \Delta + \Delta^*$ is associated with the presence of a gap in the electron density distribution, and is retained up to the critical temperature (Fig. 1b). Thus, although the S — S* system only allows the characteristics of two superconductors to be determined simultaneously, it has a number of undoubted merits over the S — n system.

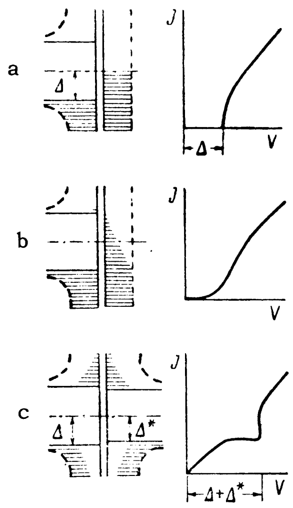


FIG. 1. Tunnel junction between a – a normal metal and a superconductor at $T = 0$; b – metal and superconductor at $T > 0$; c – two superconductors at $T > 0$. On the left the energy distribution of electrons is shown, and on the right the J-V characteristics. The line with alternate dots and dashes denotes the limiting Fermi energy; the broken line shows the density of states, the shaded portions of which show states filled by electrons. The tunnel current is determined by the transition of electrons from filled states to empty.

Tin in the form of a film less than 10^{-5} cm thick was chosen as the superconductor S* in the present work. As shown previously, for such films

[1)]For a detailed theoretical account of the tunnel transition between two metals, see [5,6].

the spread of Δ about its fundamental value amounts to 1—3%,[2,3] the properties of the films have good reproducibility, and their critical temperature T_c when condensed on tin coincides with T_c for a bulk specimen.[7]

The principal experimental difficulty in the work was to obtain a tunnel junction on the surface of a single crystal. At first we attempted unsuccessfully to obtain a junction by using insulating layers of dielectrics or of Langmuir molecular films deposited on the tin. Further experiments showed that, because of oxidation in air, an oxide layer of such thickness is formed on a freshly prepared surface of tin, even after a few hours that there is no tunnel effect through it at all. For small oxidation times this oxide film was, however, perfectly suitable for obtaining insulating layers. The thickness of the insulating film was $\sim 10^{-7}$ cm. For such thicknesses there was a high probability of holes existing in the film and, consequently, of jumpers which reduce all the effects due to the tunnel junction. This complicated still further the solution of the problem.

The following technique was used to fabricate specimens for tunnel junctions. Single crystal specimens of tin in the form of plates of thickness $\sim$ 3 mm and width $\sim$ 15 mm were cast in molds of glass. The thickness of the mold walls was ~ 0.2 mm and no wall covering of any sort was used. Before filling, the mold and the tin were heated in vacuum for several hours at 10^{-6} mm Hg and $\sim 300°$C. The filling of the mold and the growth of the single crystal from a seed was done in vacuum. The amount of impurity in the tin was 10^{-4}—10^{-3}%.

After the single crystal had been cooled, the glass was broken off, and on the surface of the tin, which was a replica of the surface of the molten glass, were deposited strips of BF-2 glue and on top, by condensation in a vacuum apparatus, tin films of width ~ 1 mm and thickness 6×10^{-6} cm. About 15 minutes elapsed between removing the glass and placing the single crystal in the vacuum apparatus, of which 9 minutes were used for drying the BF glue in a dry atmosphere in the presence of alcohol vapor. The subject of the study was the system: single crystal tin, oxide film layer, tin film (Fig. 2).

Several systems were made from one single crystal, of which ~ 20% were suitable for study. The orientation of the specimen at the locations of the tunnel junctions under study was determined by x-rays with an accuracy of $\sim 2°$. The single crystal was set up in the x-ray apparatus optically.

The J — V characteristic and the derivative

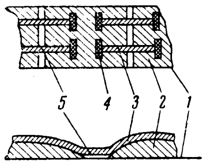
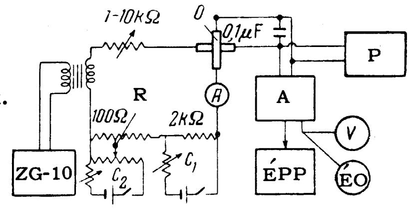


FIG. 2. Tunnel junction on single crystal specimen: 1—surface of the single crystal; 2—BF-2 layer; 3—tin film of thickness 6×10^{-6} cm; 4—Wood's alloy for soldering junction to measuring circuit; 5—the part of the specimen in which the tunnel junction exists. The tin film is separated from the bulk specimen by a layer of tin oxide.

dV/dJ (which we shall call $\mathscr{R}$ for brevity below) were measured in the course of the experiment as functions of J or V. The measuring circuit is shown in Fig. 3. The direct current through the specimen was set by the part of the circuit C_1. The circuit section C_2 was used to vary the current continuously when the recorder was writing. The current was varied by the rheostat R driven by a synchronous motor. To measure the derivative, the current through the specimen was modulated at a frequency of 87 cps. The dc voltage on the specimen was measured by the potentiometer P to an accuracy of $\sim 1\,\mu$V. The resistance of this part of the circuit was 5 k Ω. The alternating voltage was measured by the resonant amplifier A with an ÉPP-09 recorder at the output. The bandwidth of the amplifier was about 3 cps. The measurements were made with a signal of 0.5—3 μV from the specimen. The ÉO oscillograph and the voltmeter V served to monitor the ac operation of the circuit. The capacitor shunting the specimen served to reduce noise pickup.

FIG. 3. Measuring circuit.



RESULTS OF THE MEASUREMENTS

More than 100 specimens of different orientations were studied.[2)] The resistance of the specimens in the normal state was 0.1—10 Ω. The measurements were made at 1.36°K. The char-

[2)]The orientations of the studied specimens are designated on the main portion of a sphere drawn in equiangular conical projection, on Fig. 7a. The point with coordinates (θ; ϕ) = (0; 0) corresponds to the normal to the plane of the specimen lying along [001]; that with coordinates (90; 0) to the normal along [100]; that with coordinates (90; 45) to this direction along [110].

acteristics of one of the specimens studied are given in Fig. 4. They are close to what is implied by the theory of the tunnel effect between two superconductors. One difference consists of the presence of a sudden increase of current at $eV \sim \Delta$. This effect, clearly displayed in the $dV/dJ \equiv \mathscr{R}$ curve, was described by Taylor and Burstein.[8] In our experiments it is observed only in specimens with resistances less than $1\,\Omega$. The characteristics of junctions in this voltage range can be related to the tunnelling transition of "paired" electrons, and they will not be considered further. We shall be interested in the region of the sharpest increase of current near $eV = \Delta + \Delta^*$. We shall use the $\mathscr{R}(V)$ characteristics, which allow more complete information to be obtained.

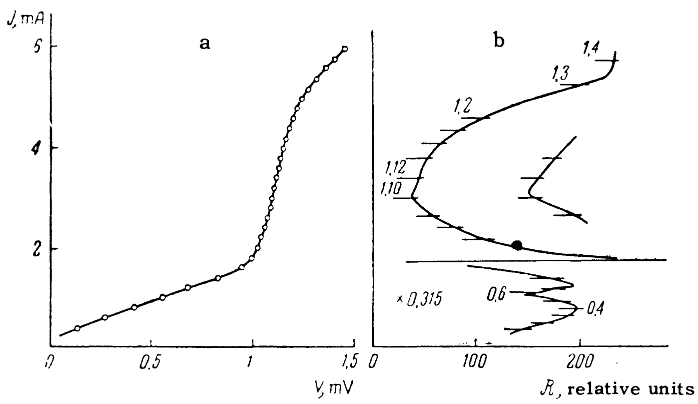


FIG. 4. Static J-V characteristic and copy of the record of $dV/dJ \equiv \mathfrak{R}$ for a tunnel junction between a tin film and a single crystal specimen for which the direction of the normal was $(\theta;\ \phi) = (40°;\ 23°)$. On the record the horizontal lines are voltage marks (mV). When recording the curve for $V < 1$ mV the modulating signal was reduced by a factor of 3.15.

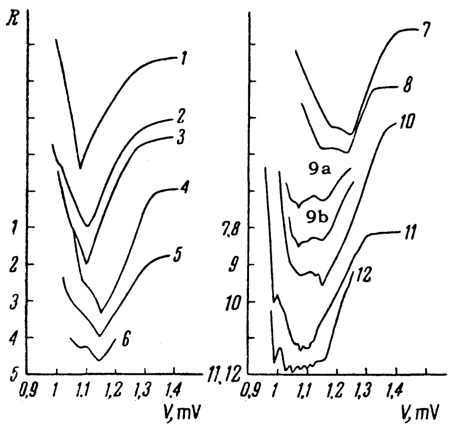


FIG. 5. Copy of the record of $\mathfrak{R}$ for specimens of various orientations: Curve 1 – $(\theta;\ \phi)$ = (54; 15.5); 2 – (82; 31); 3 – (77; 34.5); 4 – (46; 37.5); 5 – (90; 32); 6 – (90; 29); 7 – (4; –); 8 – (10; 35); 9 – (88; 19.5); 10 – (86; 18); 11 – (58; 18); 12 – (67; 6). The integers on the ordinate axis denote the zeros of the corresponding curves. Curves 6 and 2 were obtained on the same single crystal; curves 9a and 9b are records several days apart.

The specimens studied can be divided into two principal groups. To the first and larger group belong specimens with characteristics similar to those given in Fig. 4. Their distinctive feature (Fig. 5, curves 1–6), is the presence on the $\mathscr{R}(V)$ curve of a single principal minimum. The voltage V_{min} could usually be determined with error $\pm 0.5\%$ from the variation of $\mathscr{R}(V)$. For a number of specimens of this group there were discernible on the $\mathscr{R}(V)$ curve, in addition to the principal minimum, other less clearly displayed minima. However, for the majority of specimens these additional minima appear only as "wings" on the $\mathscr{R}(V)$ curve (Fig. 5, curves 4–6). In what follows we shall mainly consider the principal minimum.

To the second group (Fig. 5, curves 7–12) belong specimens with a flat minimum and specimens with two, three, or more minima close to one another in absolute magnitude on the $\mathscr{R}(V)$ curve. Of the ~ 60 specimens in this group, complicated characteristics, such as curves 11 and 12 of Fig. 5, were observed in 10 specimens.

Evidence of the reproducibility of the characteristics of a tunnel junction was provided by the following findings. The characteristics of an individual junction do not depend on the direction of change of current, and do not change with diminishing amplitude of the modulating signal. They do not change during prolonged preservation of the specimen in liquid helium or nitrogen. Storage of the specimen at room temperature usually causes the resistance of the junction to increase, but does not affect the value of V_{min} or the general form of the function $\mathscr{R}(V)$. The shape of the curve close to V_{min} does not even change when the relative size of $\mathscr{R}(V_{min})$ increases by several times due to the change in the role of "bridges" shunting the tunnel junction (Fig. 5, curves 9a, 9b). The presence of these bridges causes the characteristics of the specimen to start from some finite current at which the superconductivity of a bridge is destroyed, and the minimum on the $\mathscr{R}(V)$ curve is not so sharply displayed (9a). In fact, for such specimens a curve with a single flat minimum was usually observed instead of a curve with several discrete minima. The results obtained on specimens in which the tunnel effect was not displayed sufficiently clearly were not used in what follows.

For various tunnel junctions made on specimens of a single orientation the value of V_{min} and the general character of the $\mathscr{R}(V)$ curve agree; the relative depth of the principal minimum and the degree of resolution of the additional minima in the "wings" of the curve often differ.

The following deductions can, apparently, be made about the characteristics of junctions on specimens of neighboring orientation. In certain regions the characteristics of all specimens are similar. For example, all specimens with orientations close to (90; 45) have V_{min} = 1.16 mV; specimens with $\varphi \sim 37.5°$, $45° < \theta < 60°$ have V_{min} = 1.15 mV, etc. Along with these there exist regions where even a small change of specimen orientation ($\sim 5°$) causes the characteristics to change sharply. For example, the change from the oreintation (87; 29) to (82; 31) is accompanied by a change of V_{min} from 1.15 to 1.10 mV. This change corresponds in Fig. 5 to the transition between curves 6 and 2 obtained on one single crystal.

The way V_{min} changes when the orientation of the single crystal is changed along the path $\varphi \sim 22°$ can be traced in Fig. 6a. In the range of θ shown a sharp change of V_{min} occurs twice, close to θ = 23° and θ = 35°. In specimens with orientations between these angles, the characteristics did not vary significantly: for all specimens V_{min} $\sim$ 1.17 mV, and an additional minimum at $\sim$ 1.25 mV close to the principal in relative depth was noted on the curve $\mathscr{R}(V)$. Apparently one should ascribe specimens with identical characteristics to separate regions of orientation.

In Fig. 7a are shown the boundaries of the regions which include specimens with similar characteristics. The boundaries are shown approximately.

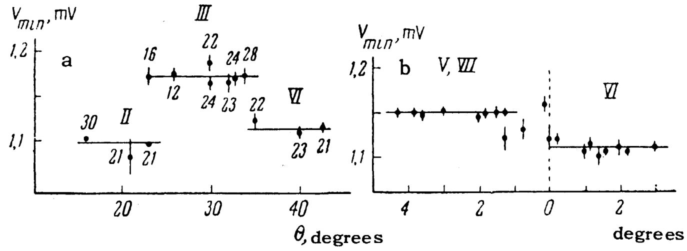


FIG. 6. a – change of V_{min} with change of specimens orientation, for orientations having values close to the path $\phi \sim 22°$. Vertical lines show the plateaux on the $\Re(V)$ curves close to V_{min}. The Roman numerals correspond to the regions of Fig. 7a. The values of the angle ϕ are given with the points; b – the change of V_{min} for specimens with orientations close to the boundaries between regions V, VII and region VI as a function of the angular distance from the boundary (the latter is shown by a broken line).

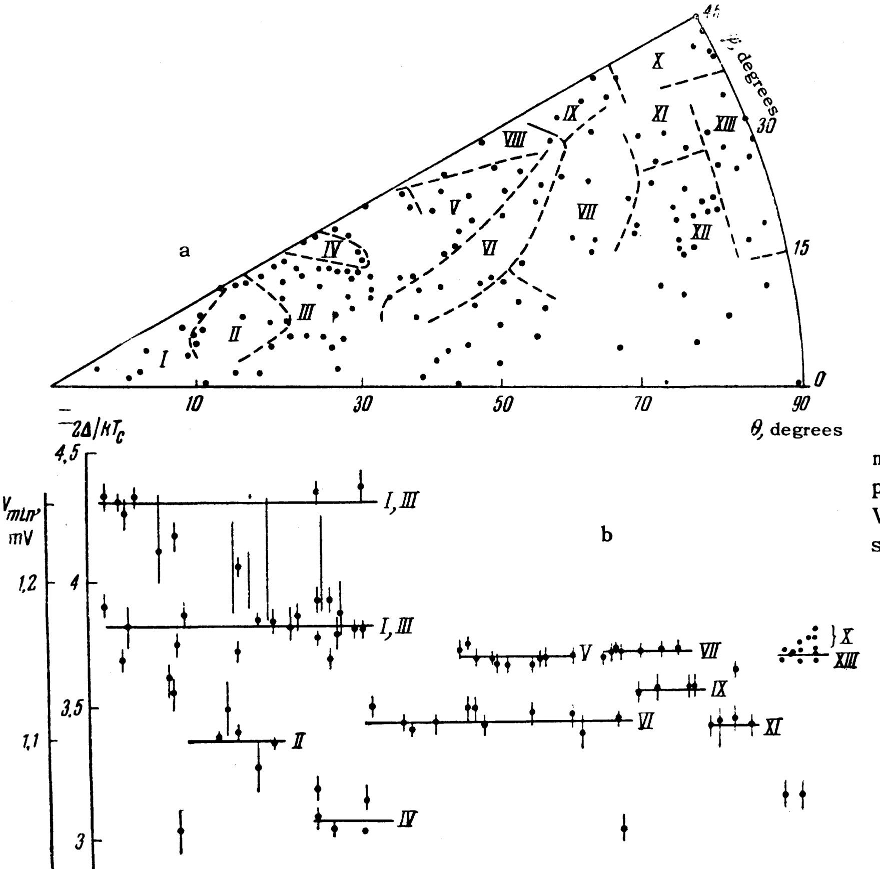


FIG. 7. a – the orientation of the specimens studied plotted on a conical equiangular projection of a sphere. Regions of constant V_{min} are shown; b – the values of V_{min} for the specimens studied.

The change of V_{min} close to the boundary of one of these regions is shown in Fig. 6b. The values of V_{min} are given in Fig. 7b for most of the specimens studied. All specimens are plotted in terms of the values of the angle θ irrespective of the value of φ. The horizontal lines are drawn through the values of V_{min} for the majority of specimens in one region.

Regions I and III of Fig. 7 include specimens with two values of V_{min} close in relative magnitude—at 1.17 and 1.25 mV (Fig. 5, curves 7, 8).

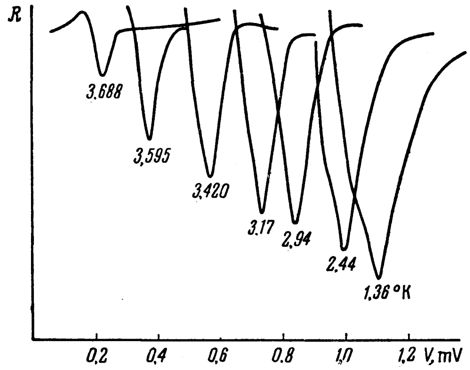
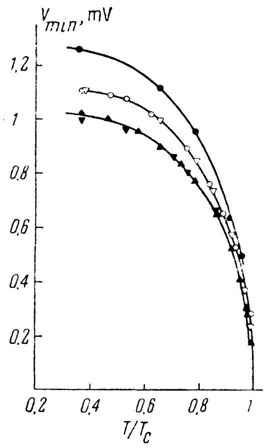


FIG. 8. The change of the $\Re(V)$ curves with temperature.

Both values of V_{min} are shown in Fig. 7b. For specimens with $\theta \sim 5°$ the deepest minimum is at 1.25 mV and for specimens with θ close to 30° it is at 1.17 mV. In a number of specimens in this region a single wide flat minimum, shown in Fig. 7b by one vertical line, is observed instead of two discrete minima. In all specimens of regions I and III, the minimum on the $\mathscr{R}(V)$ curve is wider than in most specimens of other regions. Also shown in Fig. 7b are specimens with orientations on the boundary between regions I, III and II, for which usually $V_{min} < 1.17$ mV.

Figure 7b does not show the characteristics of most specimens with orientations close to the boundaries of regions V, VII and VI in the range $30 < \theta < 60°$, which are shown in Fig. 6b. Of the specimens in region VIII ($V_{min} \sim 1.04$—1.08 mV, characteristics ill-defined), in Fig. 7b is shown only a specimen with orientation (66; 40) with two minima—at 1.04 and 1.11 mV, of which the first is the deepest. This specimen is tentatively allotted to the boundary of region VI. The specimens of region XII are not shown in the figure. The characteristics for specimens in this region at $\varphi < 28°$ are complex curves with two minima—at ~ 1.135 and ~ 1.06 mV—which merge into a single wide minimum in a number of specimens. The minimum at ~ 1.06 mV is also observed for specimens of region XIII (Figs. 4, 9, 10), for which $\varphi < 28°$.

FIG. 9. The variation of V_{min} with temperature for various specimens.



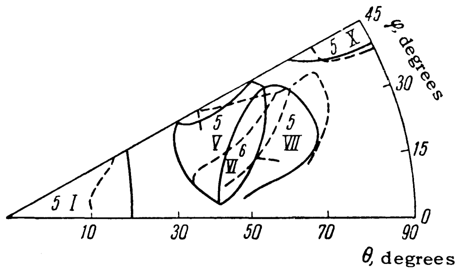


FIG. 10. Comparison of the angular dimensions of the fifth and sixth Fermi bands of tin derived from the almost free electron model (continuous lines) and regions of Fig. 7a (boundaries shown as broken lines).

The experimental data are not at present sufficient to clarify the variation of the tunnel effect with orientation outside the regions marked on Fig. 7a. Apparently, regions V and VII are bounded on the side of smaller θ by regions of smaller V_{min}. For specimens with orientations $\theta > 50°$, $\varphi < 10°$, complicated characteristics are observed similar to those given in Fig. 5, curve 12. In the region $\varphi < 10°$, $35 < \theta < 50°$ the characteristics are simple, with a single minimum at 1.13 and 1.05 mV.

A number of specimens were also studied at higher temperatures. The functions $\mathscr{R}(V)$ disappeared only above the transition temperature of tin into the normal state $T_c = 3.73°$K. The change of V_{min} is clearly caused by the variation with temperature of the gap width for tin. Similar results were also obtained for other specimens studied. The variation of V_{min} with T/T_c is given for a number of specimens in Fig. 9.

For specimens with complicated characteristics (Fig. 5, curves 9—12) the number of discrete minima resolved gradually diminishes as the temperature increases, and close to T_c only a single wide minimum is observed. In the temperature region in which the separate minima are resolved, their variation with temperature agrees with that obtained on specimens with simple characteristics.

DISCUSSION OF THE RESULTS

The method of study adopted only allowed us to determine simultaneously the characteristics of a single crystal of tin and a thin film condensed on its surface. However, the following facts allow us to conclude that the change of the $\mathscr{R}$(V) characteristics is caused by the properties of the single crystal tin: 1) data on the reproducibility of the properties of tin films;[7] 2) the agreement between the characteristics of tunnel junctions fabricated in different experiments on single crystals of neighboring orientations; 3) the difference between the characteristics of tunnel junctions obtained in a single experiment with a single film but on differently oriented parts of a single crystal.

The type of change with temperature of the $\mathcal{R}(V)$ curves, and particularly the disappearance of the effects associated with the tunnel junctions at T_c, not to mention the variation with the magnitude of V_{min}, show that the phenomena observed cannot be explained by the formation on the surface of the metal by any hypothetical cause of regions with different values of T_c. The results obtained should, it seems, be associated with the anisotropy of Δ for a single crystal of tin.

In order to transfer from the anisotropy of V_{min} to the anisotropy of Δ for tin, it will be supposed that $eV_{min} = \Delta + \Delta^*$. This is to a certain extent an assumption. In the tunnel effect between two superconductors one would expect, according to the theory, the current to rise sharply and, thus, $\mathcal{R}$ to become zero at $eV = \Delta + \Delta^*$. It is, however, obvious that in a real system there are enough causes for the jump to be smeared out and $\mathcal{R}$ to be increased. One such reason is, for example, the non-ideal properties of the tin films, in which a similar difference between calculation and experiment was first discovered.[2,3] For calculation we use $\Delta^* = 0.56$ mV.[2] for tin films. The error in this value of Δ^* does not exceed 2—3%. Correspondingly, a similar systematic error is also possible in Δ for bulk tin.

The values of V_{min} and $2\Delta/kT_c$ for tin are compared in Fig. 7b. The smallest value of V_{min}, amounting to ≈ 0.99 mV, observed for a number of specimens with complicated characteristics (see, for example, Fig. 5, curves 11, 12), corresponds to $2\Delta/kT_c = 2.7$; the largest, $V_{min} = 1.25$ mV, corresponds to the value $2\Delta/kT_c = 4.3$. Thus, depending on the crystallographic orientation of the specimen, the gap width of tin changes by a factor of 1.5. The anisotropy of Δ for tin is very complicated. Large regions of approximately constant Δ and sharp boundaries between them are, apparently, its distinctive feature. The discovery of sharp boundaries (Fig. 6b) was the most unexpected result of the measurements. It was difficult to suppose a priori that the surface of a metal, even when it is a replica of molten glass, possesses sufficient smoothness to enable one to observe a change of V_{min} corresponding to a change of orientation of $\sim 5°$.

Such a peculiar type of anisotropy in Δ is probably associated with peculiarities in the electronic structure of tin. The sudden change of Δ at the boundary between regions (Fig. 7a) can be explained by the fact that for specimens of neighboring regions the principal contribution to the tunnel current is provided by electrons from different Fermi bands of tin, the gap width in which can differ.[9,10] The small change of Δ within each of the regions is caused by the weak anisotropy of Δ throughout the band, which is a second order effect compared with the anisotropy both of the electronic spectra and of the electron-phonon interaction.[11-13]

We compare the results we have obtained with data on the Fermi surfaces of tin. A number of workers[14-17] have described experimental studies of this problem; however, the structure of all the bands is not yet unravelled. It has only been shown reliably that some sections of the fourth hole band approximate to the predictions of the almost free electron model. We shall use this model for comparison in the first approximation. The major contribution to the tunnel current is carried by electrons with $v = \partial E/\partial p_n$, where p_n is the momentum normal to the junction plane. We shall determine the directions of $\partial E/\partial p_n$ for electrons of various bands, or, what is equivalent, the directions of the normals to the constant energy surfaces in momentum space. This problem is easily solved by the geometrical construction method,[18] for which we use the fact that for tin $a/c = 1.83$ and the radius of the free electron sphere is $r = 1.52\,(2\pi/a)$. The results of the construction for part of the fifth band and one of sixth bands of tin are shown in Fig. 10. For comparison regions I, VI, V, VII, and X from Fig. 7a are plotted on the same diagram. If it is assumed that regions I, V, VII and X with a similar value of Δ correspond to the fifth band, and VI to the sixth, then some correlation can be seen in both the relative position and angular dimensions of the Fermi bands and the regions of constant Δ. Such a correlation can also be traced for certain other regions.

A complete coincidence of the boundaries is not, however, observed. Furthermore, in clear contradiction to the almost free electron model, regions with complicated characteristics are found, the appearance of which indicates the participation in the tunnel current of several groups of electrons with different gap widths. This discordance can, it seems, be associated with distortion of the Fermi surfaces in a real crystal relative to those constructed geometrically. Such a distortion is most probable close to where the bands intersect the boundaries of the unit cell of the reciprocal lattice and where the separate pieces of a band are joined together, due to the rounding off of sharp angles. The latter, for example, apparently corresponds to an increase of the angular dimensions of the bands, which can cause them to overlap. When such overlapping of bands occurs it is natural to expect complex $\mathcal{R}$ characteristics to appear. The side wings on the $\mathcal{R}(V)$ curves can also probably be related to rounding off of band corners. Deviations from the almost free electron model have also been discovered by direct experimental measurement of the dimensions of the Fermi bands of tin. For example, according to Gantmakher's data,[17] important deviations from the almost free electron model occur in both the third and fourth bands. Unfortunately, there is at present no direct detailed study of the fifth and sixth bands of tin.

Thus, the experimental results, taken as a whole, do not contradict the supposition that neighboring regions with differing values of Δ do, in fact, correspond to different bands of the Fermi surface of tin. It should, however, be emphasized that the experiments carried out did not prove the uniqueness of the interpretation of the experimental data which has been given above.

When comparing our results with data on the anisotropy of tin obtained from ultrasonic absorption[19] or high frequency field[20] experiments, one should take into account that both these methods give only an average feature of the anisotropy. The type of averaging is determined by the model of a superconductor with which comparison is made. For example, from the ultrasonic absorption in a superconductor with one band, only the value of Δ_{min} for directions perpendicular to the propagation of sound is determined. Thus, on the basis of the data obtained one would expect for propagation along [110], as well as for most other directions, the values $2\Delta/kT_c = 3.0$, while direct measurements give $2\Delta/kT_c = 3.8$. If a model with several bands is used for the superconductor a more complex averaging is required, for which the characteristics of all bands are needed. If it is supposed that the contribution of each band of constant Δ is proportional to its angular size, then we obtain for the [110] direction $2\Delta/kT_c = 3.7$. However, the agreement of this value with the experimental data is perhaps accidental also.

We now turn to results obtained at temperatures above 1.36°K. They allow the temperature variation of the gap width for tin to be determined. The data obtained are shown in Fig. 11. For all the values of Δ studied the variation of $\Delta(T/T_c)$ is close to that predicted by the present theory of superconductivity. The maximum deviation from the theoretical variation does not exceed 3–5%.

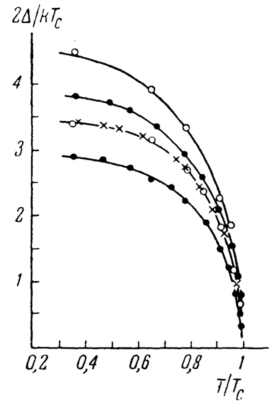


FIG. 11. The variation of the gap width of tin with relative temperature T/T_c.

The change with temperature of the relative anisotropy lies within the same limits. The latter result is in agreement with the analysis made by Pokrovskiĭ[12] of the variation with temperature of the anisotropy of a superconductor.

The author is grateful to P. L. Kapitza for interest in the work and support, and to A. I. Shal'nikov for valuable critical remarks and discussion.

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Translated by K. F. Hulme
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