STUDY OF TIN BY THE TUNNEL EFFECT METHOD. II

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The width of the gap Δ in the electron energy spectrum of a single crystal of tin in the superconducting state is studied by the tunnel effect method. The complicated nature of the anisotropy of Δ is associated with the presence of surfaces along each of which Δ varies slightly. Surfaces, reconstructed according to the experimental data, are compared with the Fermi surfaces of tin.

As a result of a study by the tunnel effect method of the width of the gap Δ in the electron energy spectrum of tin,[1] a significant variation of Δ, depending on the crystallographic orientation of the sample, was observed. The anisotropy of Δ is of a complicated type. Large regions of approximately constant Δ and sharp boundaries between them are, apparently, its distinctive features. In order to explain the results, it was proposed that the complicated nature of the anisotropy is associated with singularities of the Fermi surfaces of tin. In order to verify this hypothesis, it would be necessary to try to reconstruct, according to experimental data, the surfaces along which the width of the gap varies insignificantly, and then compare them with the Fermi surfaces of different bands. However, the data obtained turned out to be inadequate for the reconstruction of these surfaces. Previously[1] it was only possible to observe in certain sections a correlation in the distribution of regions of constant Δ and of the Fermi surfaces constructed according to the model of nearly free electrons. In this article, a more detailed analysis of the data on anisotropy of the gap is carried out, based on the results of further investigations.

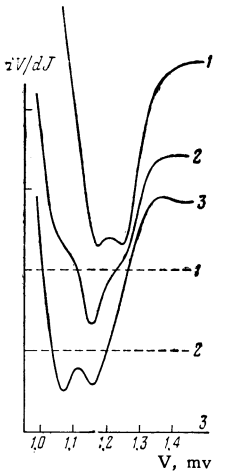

FIG. 1. Copy of the $dV/dJ \equiv \mathcal{R}(V)$ curves for tunneling transitions for samples of various orientations: curve 1 – $(\theta; \phi) = (2°; 0)$, curve 2 – $(\theta; \phi) = (45°; 17°)$, curve 3 – $(\theta; \phi) = (77°; 10°)$ (for convenience, the origin has been displaced along the axis of ordinates for different curves).

1. METHOD OF MEASUREMENT AND RESULTS

The width of the gap Δ, just as in[1], was determined from the characteristics for the tunneling transition through an insulating layer $\sim 10^{-7}$ cm between a single crystal of tin and a tin film of thickness $\sim 6 \times 10^{-6}$ cm. The current-voltage characteristics of the transition and the derivative $dV/dJ \equiv \mathcal{R}(V)$ were recorded in experiments at 1.36°K. In contrast to the case of a tunneling transition between two thin films, the curve $\mathcal{R}(V)$ for a single crystal has a more complicated shape—usually with several discrete minima. These minima may be similar in regard to relative depth (Fig. 1, curves 1 and 3) or they may differ by several times. In the latter case they appear as "wings" on the curve of the principal minimum (Fig. 1, curve 2).

The complicated characteristics of tunneling transitions are evidently due to the fact that several groups of electrons in a single crystal, with different gap values Δ_k, contribute to the tunneling current. In order to calculate Δ_k, it was assumed that the voltage corresponding to a minimum of the curve $\mathcal{R}(V)$ is equal to $eV_{min} = \Delta_k + \Delta^*$, where $\Delta^* = 0.56$ meV is the width of the gap in the electron energy spectrum for a tin film. In what follows, the gap Δ_k will be given in relative units: $(\Delta_k) = 2\Delta_k/kT_c$.

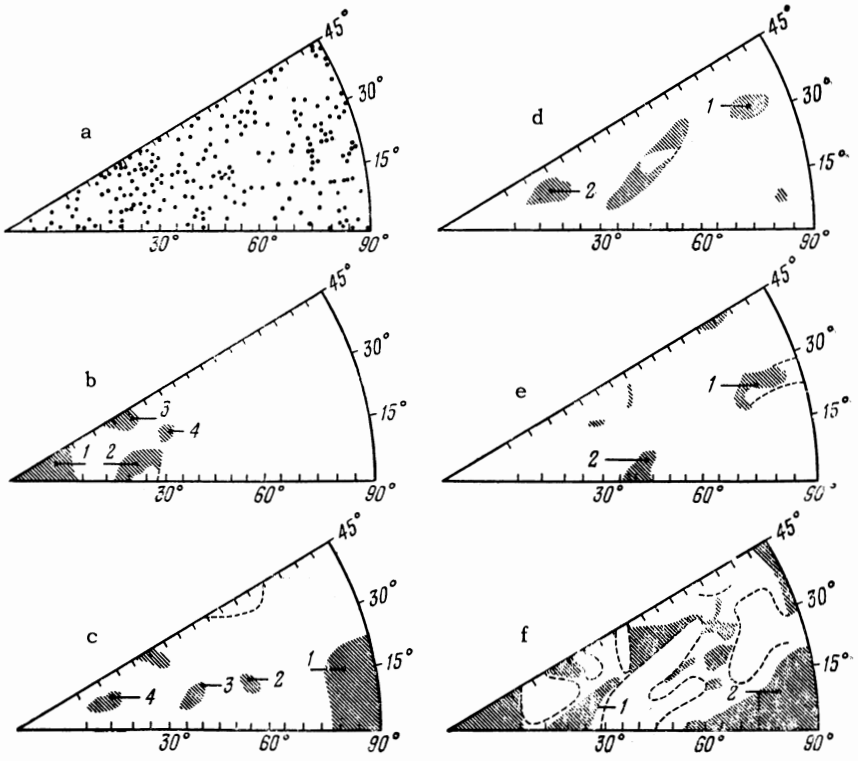

FIG. 2. a – the orientation of the samples studied plotted on a conical equiangular projection of a sphere. b through f – regions corresponding to various values of (Δ): b – the cross-hatching indicates the regions corresponding to values of (Δ) ~ 4.3; c – cross-hatching indicates the regions with (Δ) ~ 3.1, the dashed line indicates region VIII of article [1] with the assumed value (Δ) ~ 3.1; d – the cross-hatching indicates the regions (Δ) ~ 3.4, e – the regions (Δ) ~ 3.55; f – the cross-hatching indicates the regions with (Δ) = 3.7 to 3.8, the dashed lines indicate the boundaries of the regions in which the samples, whose $\mathcal{R}$(V) curves do not have a minimum at (Δ) ~ 3.7 to 3.8, are located.

The main portions of the sphere for tin are represented on Fig. 2 in an equiangular conical projection. The point with coordinates (θ; φ) = (0; 0) corresponds to the direction of the normal to the plane of the sample lying along [001]; the point (θ; φ) = (90°; 0) corresponds to the direction of the normal along [100]; the point (θ; φ) = (90°; 45°) corresponds to direction of the normal along [110]. The orientations of the samples which are utilized in this communication are indicated on Fig. 2a.

Let us observe how samples with similar values of Δ_k are distributed. As already indicated, the tunneling current is usually caused by several groups of electrons with different values of Δ_k. The contribution of each group can be determined from the depth of the corresponding minimum. The most intense groups correspond to very deep minima. In what follows, we shall consider primarily these minima.

Depending on the crystallographic orientations, the width of the gap for a single crystal of tin varies up to 1.5 times: from $(\Delta_k) \approx 4.3$ to $(\Delta_k) \approx 2.8$; in this connection, for the majority of samples $(\Delta_k) = 3.7$ to 3.8. It is obvious that, because of the finite width of the minima on the complicated $\mathcal{R}$(V) curves, it is only possible to determine accurately the value of Δ_k for substantial differences in the values of Δ.

In the first place, let us consider samples with a minimum at 1.24 to 1.26 meV [(Δ) = 4.25 to 4.35], which is clearly indicated on the curves of $\mathcal{R}$(V) (Fig. 1, curves 1 and 2). If we single out the samples which have one of the deepest minima located at (Δ) ~ 4.3 (for example, a sample which corresponds to curve 1 in Fig. 1), then it turns out that all of them are grouped in a few regions. In what follows we shall refer to these regions as "regions of corresponding values of Δ." The regions with values of Δ between 4.25 and 4.35 are indicated on Fig. 2b. The boundary of the regions is determined, in the case of a sufficiently large number of studied samples, as the boundary a transition through which is accompanied by an abrupt change of the relative depth of the corresponding minimum on the tunneling transition characteristics (see[1]). Thus, for samples located away from the boundaries of the regions of Fig. 2b by an angular distance of a few degrees, the depth of the minima on the $\mathcal{R}$(V) curves for (Δ) ~ 4.3 is already smaller than the basic depth by a factor 2 or 3. However, some uncertainty in the determination of the boundaries arises because of the finite number of samples studied. For example, because of the absence of samples near $\varphi = 0$, $20° < \theta < 25°$ (Fig. 2a), it is impossible to assert with complete certainty that region 2 of Fig. 2b extends to the $\varphi = 0$ axis, as indicated on the figure by the dotted line.

We note that all of the regions with values of (Δ) ~ 4.3 are located near the [001] axis in an inter-

val ~ 35°. For some of the samples located outside of this interval, the minimum associated with $(\Delta) \sim 4.3$ appears in the form of a "wing" on the $\mathcal{R}(V)$ curve (see, for example, curve 2 on Fig. 1).

The regions with values of (Δ) between 3.05 and 3.25 are indicated on Fig. 2c. In the angular interval $\theta > 35°$, (Δ) has a value from 3.15 to 3.25; in the interval $\theta < 35°$, we have $(\Delta) = 3.05$ to 3.15. This value of (Δ), in the form of a "wing" on the curve, is observed in the same way for all samples lying in the region $\theta < 45°$. We note that $(\Delta) = 3.05$ to 3.25, as well as $(\Delta) = 4.25$ to 4.35, is observed in the majority of regions in the presence of $(\Delta) = 3.7$ to 3.8 (Fig. 1, curves 1 and 3).

The regions with $(\Delta) = 3.4$ are indicated in Fig. 2d. None of these regions, with the exception of those lying near the point $(\theta; \varphi) = (85°; 9°)$, contain the value $(\Delta) = 3.7$ to 3.8.

For a number of the samples studied, the principal minimum on the curve corresponds to $(\Delta) = 3.53$ to 3.58. This value of Δ is too close to $(\Delta) = 3.7$ to 3.8 to be able to distinguish all regions. The reliably-established regions are indicated on Fig. 2e.

The region with values of (Δ) between 3.7 and 3.8 is indicated on Fig. 2f. The value $(\Delta) = 3.7$ to 3.75 predominates in the angular interval $\theta > 35°$, and the value $(\Delta) = 3.8$ in the interval $\theta < 35°$. The boundaries inside of which the principal minimum on the $\mathcal{R}(V)$ curves is not present when $(\Delta) = 3.7$ to 3.8 are indicated by dashed lines on Fig. 2f.

Although a large number of samples were used for the construction of Fig. 2, the boundaries of a number of the regions are not determined very precisely. The point is that, because of the extremely complicated nature of the anisotropy of Δ for tin, we assumed it possible to combine into one region of Fig. 2 adjacent samples with the same value of Δ only in that case when their orientations differed by less than 5°, although in certain cases, for example, the long region $(\Delta) = 3.4$ on Fig. 2d, this requirement was obviously too stringent. Because of this we do not consider, in particular, the value $(\Delta) = 2.8$, observed only for certain samples. However, even in the most favorable case, the accuracy of the determination of the boundaries is limited by errors in the determination of the orientation of the samples, which may exceed 2°.

2. DISCUSSION OF RESULTS

Let us consider the obtained data from the point of view of the hypothesis introduced in[1]. According to this hypothesis, the complicated nature of the anisotropy of Δ is associated with the fact that electrons belonging to several bands of tin, with different gap widths, give a contribution to the tunneling current. In a given direction, the electrons with $v = \partial E/\partial p_n$, where p_n is the momentum normal to the plane of the transition, give the principal contribution. The relative contribution of two bands in a selected direction is proportional to the radius of curvature of the Fermi surface in each of the bands.

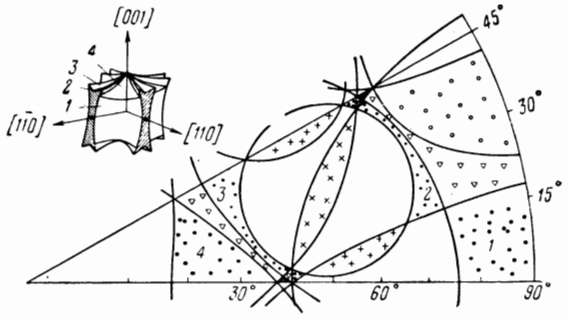

FIG. 3. Regions for the directions of the normals to the Fermi surfaces of tin, constructed according to the nearly free electron model. Crosses – ■ second band; ▽ – third band; ●, ○ – fourth band; × – the two sixth bands; the fifth band remains without any identifying marks. In the upper left hand corner, there is a figure showing the fourth "hole" band.

For instance, let us consider what kind of results would be expected if the Fermi surface of tin were to coincide exactly with the model of nearly free electrons. For this purpose, let us determine the value of $\partial E/\partial p_n$ in different bands, or, what is equivalent, the direction of the normals to the surfaces of constant energy in momentum space. This problem is easily solved by the geometrical construction method,[2] for which we assume $a/c = 1.83$ and a free electron sphere radius $r_0 = 1.52(2\pi/a)$. The results of the construction are shown in Fig. 3. The boundaries between bands are the intersections of the free electron spheres with a system of planes parallel to [001] and one of the two other principal axes, and of planes situated at angles of 61° 22′ and 39° 16′ to the [001] direction (the trace from the latter plane is seen on Fig. 3 in the form of a nearly perfect circle). The Fermi surface of the metal, according to the free-electron model, is in fact constructed out of the regions of Fig. 3, as depicted in the upper corner of Fig. 3 for the hole surface of the fourth band. According to the stated hypothesis, the boundaries on which an abrupt change of Δ occurs should coincide with the boundaries shown in Fig. 3.

Although some correlation was previously[1] noticed for $\varphi > 15°$ in the location of the sixth and

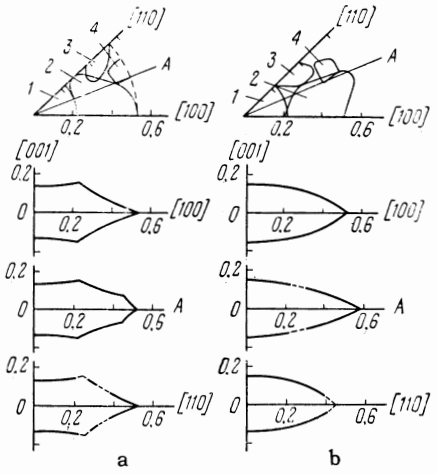

FIG. 4. Reconstructed "hole" (a) and "electron" (b) surfaces for $(\Delta) = 4.3$. The upper figure is a projection of the regions with values of $(\Delta) \sim 4.3$, from which the surface in the (001) plane is constructed; the lower figures represent several cross sections of the surface. The numbering of the regions is the same as in Fig. 2b. Distances along the axes are measured in units of $(2\pi/a)$.

fifth bands and of the regions $(\Delta) = 3.4$ and $(\Delta) \sim 3.75$, it is obvious that complete matching of the regions represented in Figs. 2 and 3 does not occur. And what is more, regions with complicated characteristics are found, in clear contradiction with the nearly free-electron model. These discrepancies are apparently associated with a substantial distortion of the Fermi surfaces in a real crystal relative to those constructed geometrically according to the free-electron model. Before making a detailed comparison of the obtained data with experimental studies of the Fermi surface of tin, it is clear that one should verify whether the results obtained are not in clear contradiction with the hypothesis stated above, that is, whether from the regions of Fig. 2 with close values of Δ one can construct closed surfaces, similar to the way in which bands are constructed out of the pieces of Fig. 3. (We recall that, according to theoretical calculations[3,4], there is a very small probability for any substantial variation of Δ along one band.) Only after this has been done can we attempt to compare the obtained surfaces of constant Δ_k with data about the Fermi surfaces of tin.

In order to construct each of the surfaces of constant Δ_k, we have available only information with regard to its angular dimensions, more precisely, the angular dimensions, evidently, of its flattest sections. We shall assume that the radius of curvature of all of these sections is the same. This is an additional assumption since, according to the conditions of the experimental arrangement, it is impossible to determine the dependence of a change in the absolute depth of a minimum on crystallographic direction for a selected value of Δ_k. For definiteness, let us assume that the radius of curvature of all the surfaces coincides with r_0—the radius of the free electron sphere. It is obvious that, as in the case of bands constructed according to the nearly free-electron model, each of the closed surfaces consists either only of pieces of spheres with the radii directed inside the surface, or only of pieces with the radii directed outward. According to conventional terminology, in the first case we shall call the surface "electron," and in the second case—"hole."

The results of constructions of two possible versions of the surface corresponding to $(\Delta) \approx 4.3$ are shown in Fig. 4. Although the experimental data are in better agreement with the "hole" surface version, there is not enough data to make a final selection. As is evident from Fig. 4, the surfaces of both constructions do not differ substantially. In the first approximation they remind us of a lens, whose ratio of diameter to height is 3:1.

In the course of the construction, we neglected the angular regions in which the minimum for $(\Delta) \approx 4.3$ is several times less than the principal minimum and appears only as a "wing" on the $\mathcal{R}(V)$ curve (Fig. 1, curve 2). In these regions of the surface $(\Delta) \sim 4.3$, the radius of curvature is evidently several times smaller than for the sections indicated on Fig. 2b. The linear dimensions of these regions are accordingly smaller by the same factor. According to estimates, the total contribution from all of these regions does not exceed the errors associated with the inexact determination of the boundaries on Fig. 2b.

Constructions were also carried out for other values of Δ_k. The surface $(\Delta) \sim 3.1$ is an "electron" central surface, somewhat flattened along the {001} and {100} axes (Fig. 5a). It is possible to group the three regions of values of $(\Delta) \sim 3.4$ into only two closed surfaces. The two regions indicated by numerals on Fig. 2d form a small central surface. It is impossible to determine with certainty whether this is an "electron" or "hole" surface. A more probable version of the "electron" surface is shown in Fig. 5b. The large region in Fig. 2d which is not labelled with numerals, together with the similar regions in the other quadrants, form four elongated surfaces per unit cell. These surfaces are similar to the large surfaces of the sixth zone, constructed according to the free-electron model. Thus, for both of the surfaces compared, the ratio of the major axis to the smallest is 3:1, the length of the major axis amounts to $\sim 0.95(2\pi/a)$. However, the orientation of these surfaces to the lattice is somewhat different. Whereas the major axes of the surfaces for

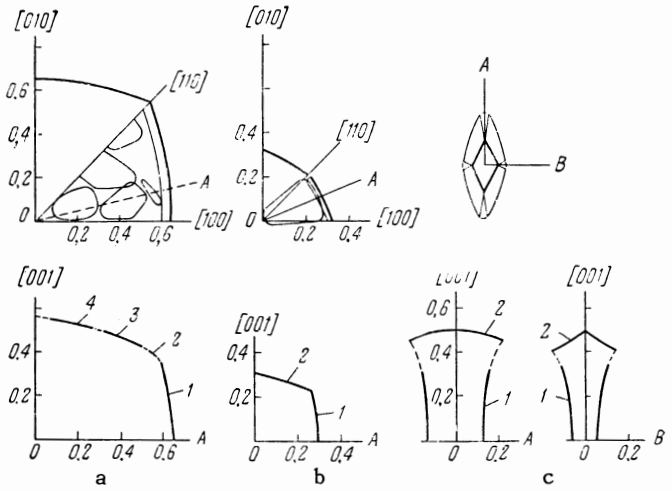

FIG. 5. a – the surface (Δ) ~ 3.1; b – one of the surfaces (Δ) ~ 3.4; c – the surface (Δ) ~ 3.55. The numbering of the regions is the same as in Fig. 2. Distances along the axes are measured in units of $(2\pi/a)$.

the nearly free electron model lie in the {100} planes, making an angle of 61° 22′ with the [001] axis, for the surfaces with (Δ) ~ 3.4 they make an angle ~ 20° with the {100} planes and an angle ~ 50° with the [001] axis.

Only the regions of largest dimensions (indicated by numerals in Fig. 2e) were used for the construction of the surface (Δ) ~ 3.55. These regions are grouped together in two noncentral "hole" surfaces per unit cell. Several cross sections of the surface (Δ) ~ 3.55 are shown in Fig. 5c.

We turn, finally, to the regions of values (Δ) = 3.7 to 3.8. These regions occupy such an extensive angular interval that if we attempt to join them together into one surface, similar to what we did earlier, then its geometrical dimensions in all directions will considerably exceed the dimensions of the unit cell. This may signify that either the radius of curvature of the surface is less than r_0 or else these values of Δ correspond to several isolated surfaces, as in the case (Δ) ~ 3.4. Since it is difficult to presuppose that one of the most extended, very deep minima of the $\mathcal{R}(V)$ curves is associated with the surface having the smallest curvature, the first assumption seems very unlikely to us. It is more probable that these values of Δ are related to several surfaces, perhaps those corresponding to the fifth and fourth "hole" bands of the free-electron model. Some correlation in the distribution of these bands on Fig. 3 and the regions of values (Δ) = 3.7 to 3.8 on Fig. 2f testifies in favor of this supposition. And what is more, if it is assumed that regions 1 and 2 of Fig. 2f refer to the fourth band, but all other regions pertain to the fifth band, then the constructed surfaces agree with the fourth and fifth bands of the free-electron model in all of its basic dimensions. It should be emphasized, however, that such a separation of the regions into two surfaces is, to a considerable extent, arbitrary.

Thus, although the regions corresponding to a definite value of Δ are, at first glance, randomly distributed, they are all joined together into closed surfaces of a basically simple geometrical shape. Some of these surfaces are similar to the Fermi surfaces of tin, constructed according to the nearly free electron model; however, other surfaces, for example, (Δ) ~ 4.3 and (Δ) ~ 3.1 differ markedly from this model.

Now let us compare the obtained data with experimental studies of the Fermi surface of tin. A large number of articles[5-9] have been devoted to this problem. The investigations[7-9], in which direct measurements of the geometrical dimensions of the cross sections of the Fermi surfaces were made, are of most interest.

First, let us turn to investigations carried out by the high-frequency method.[8,9] In these studies it was reliably established that there exists a surface in tin which is identical, with respect to its own basic dimensions, with the fourth "hole" surface of the nearly free electron model. As already indicated above, it is possible that one of the (Δ) ~ 3.75 surfaces coincides with it. Furthermore, in these studies it is shown that a surface similar to the third "hole" surface exists in tin. The geometry of this surface is similar to the surface (Δ) = 3.55. Moreover, in this case there is even an astonishing agreement of the geometrical dimensions. Thus, the cross sectional area of the (Δ) ~ 3.55 surface in the (001) plane amounts to 0.01 to $0.02(2\pi/a)^2$, whereas the cross sectional area of the Fermi surface of tin, according to[5], is equal to $0.014(2\pi/a)^2$. Both the size of the surface (Δ) = 3.55 as well as the size of the surface along [001] amount to $\sim 0.5(2\pi/a)$, according to[8,9]. Similar agreement of geometrical dimensions must indicate that the radius of curvature of the surface is actually close to r_0. Aside from this, in tin the presence of a surface differing substantially from all of the surfaces constructed according to the free-electron model has apparently been established.[9] It looks like the surface (Δ) = 3.1. Its (001) cross section is identical to the cross section of the surface (Δ) ~ 3.1, although it is more flattened along the [001] axis. It is extremely probable that the value (Δ) ~ 3.1 pertains precisely to this surface. Then, since the geometrical dimensions of the surface (Δ) ~ 3.1 are larger than the dimensions of the Fermi surface, determined

by direct measurements, one would expect that the radius of curvature of the surface is less than r_0.

Unfortunately, there is not enough reliable information about the shape of the Fermi surfaces in the remaining Brillouin zones. And what is more, present high-frequency[9] and ultrasonic[7] studies in considerable measure have not been deciphered, and in a number of cases they do not agree. It would be easy to relate the averages of some of these undeciphered data to the surfaces $\langle\Delta\rangle \sim 4.3$ and $\langle\Delta\rangle \sim 3.4$; however, it is hardly appropriate to carry out a detailed comparison before completion of the experimental investigation of the Fermi surface of tin.

Thus, the totality of experimental data, about the anisotropy of the tunnel effect as well as about the peculiarities of the Fermi surface of tin, does not contradict the above-stated earlier hypothesis about the existence in this metal of a direct connection between the anisotropies of Δ and special characteristics of the Fermi surface. It is of interest to note that here, on all surfaces which do not differ qualitatively from the nearly free electron model, the gap width is close to the theoretical value $\langle\Delta\rangle \sim 3.75$. From a theoretical investigation of the simplest model of a superconductor with isotropic surfaces of two bands,[10] it follows that the gap width of the latter may differ only in the case when the electron-phonon interaction between bands differs from the electron-phonon interaction within the limits of one band. It is possible that precisely this case is realized in tin.

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