MEASUREMENT OF SURFACE TENSION AT THE BOUNDARY BETWEEN SUPERCONDUCTING AND NORMAL PHASES

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The surface tension between superconducting and normal phases has been determined by measuring the period of a simple laminar structure, obtained in a flat sample located in an inclined magnetic field. For a single crystal of tin, the results of measurements in the temperature interval 2.165° K to 3.5° K can be described by the relation $\Delta = 2.5 \times 10^{-5}(1 - T/T_c)^{-1/2}$ cm.

A theoretical investigation of the structure of the intermediate state was first carried out by Landau[1] as early as 1937. For a sample in a transverse magnetic field, the model of the structure which he proposed and regarded as the simplest was one in the form of alternating layers of superconducting and normal phases (s- and n-phases), a cross-section of which is shown in Fig. 1. The size of the layers in this model is related to the sample thickness and to the magnitude of the surface tension at the phase boundary by the expression

$$\alpha\,(8\pi/H_c^2) = (a^2/L)\,\varphi\,(\eta), \tag{1}$$

where H_c is the critical field, φ is a dimensionless function whose numerical value has been calculated elsewhere,[2] and $\eta = a_n/a$; the meaning of the remaining symbols is given in the figure. The quantity $\alpha \times 8\pi/H_c^2$, which has the dimensions of length, is usually designated as Δ.

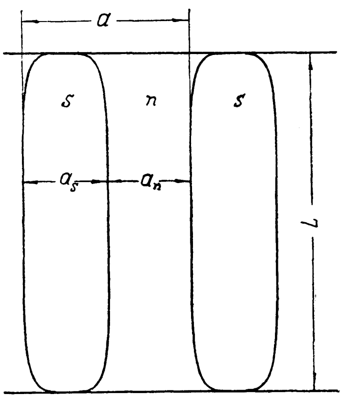

FIG. 1

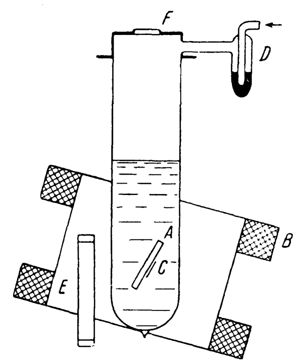

FIG. 2. Schematic diagram of the apparatus

A determination of the geometrical size of the s- and n-domains thus affords the principal opportunity for calculating the quantity Δ. An experimental investigation of the structure of the intermediate state by other methods has shown, however, that the shapes of the s- and n-domains are usually very complex, and far removed from the simple cases studied theoretically. Furthermore, the irregularity and irreproducibility of the structures has indicated that the intermediate state of the actual samples was not an equilibrium one, whereas the theoretical study presupposed thermodynamical equilibrium.

We have undertaken a search for experimental conditions under which the structure would have a simple form, permitting mathematical treatment, and for which it would be possible to obtain information concerning the degree of approximation of the structure to an equilibrium state.

Balashova,[3] while studying the structure of the intermediate state of a tin sphere by using a ferromagnetic powder, noticed that over a small region of the surface of the sphere, where the external field made an acute angle with it (i.e., near the magnetic equator of the sphere), the picture was much simpler than over the rest of the surface, and the n-domains had the form of comparatively wide, straight zones in the meridional direction, independent of the method by which the specimen was carried over to a given point of the region of the intermediate state on a diagram in the variables H and T. It was presently shown by Dzialoshinskii[4] that this alignment of the laminae is energetically the most favorable one. Proceeding from this, we have carried out a study of the structure of the intermediate state for a sample located at an acute angle β with respect to an external magnetic field.

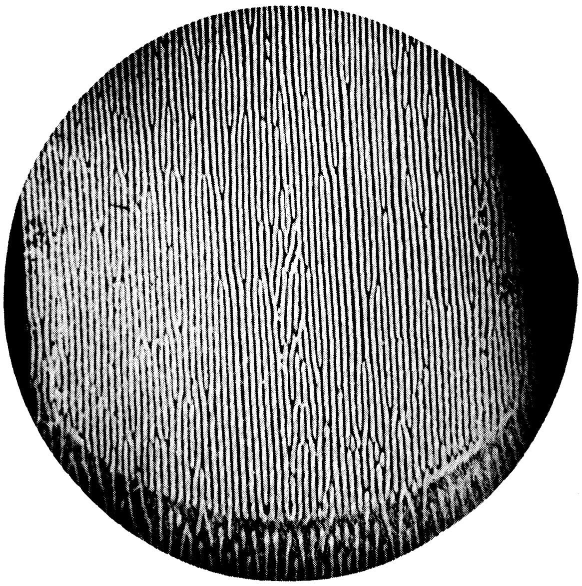

FIG. 3. View of the surface of the disc 50 mm in diameter (sample No. 1). $T = 2.165°$ K, $h = 0.950$; $\eta = 0.62$, $a = 0.54$ mm.

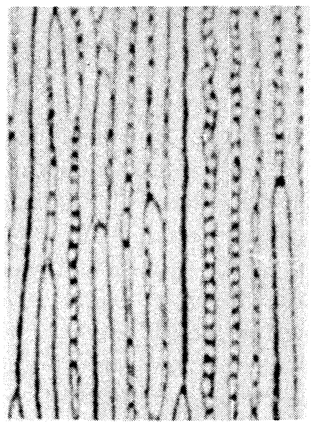

FIG. 4

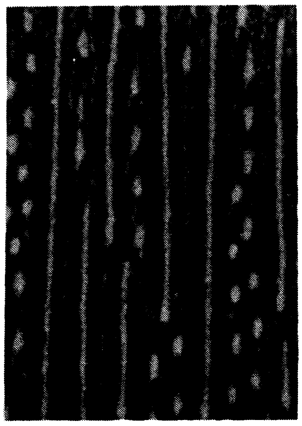

FIG. 5

FIG. 4. A portion of the surface of sample No. 1 (visual field is 7.3 mm wide). $T = 2.165°$ K, $h = 0.910$, $\eta = 0.5$.

FIG. 5. A portion of the surface of sample No. 1 (visual field is 12 mm wide). $T = 2.165°$ K, $h = 0.995$.

After preliminary experiments, we settled upon a sample in the form of a disc 50 mm in diameter and 2 mm thick, at an angle $\beta = 15°$. A schematic drawing of the experimental configuration is shown in Fig. 2. The sample A is located in the field of the coil B, fixed to an axis perpendicular to the plane of the drawing. Initially the field of the coil was directed along the surface of the sample, which was ascertained from a maximum in the conductivity of a thin tin film C, mounted parallel to the surface of the sample, after which the coil was turned through an angle β. The accuracy of the setting amounted to several minutes. The field of the earth was compensated by Helmholtz coils, not shown in the figure.

The structure of the intermediate state was exhibited with the aid of a fine nickel powder, blown into the Dewar flask from the atomizer D.[3] The surface of the sample was illuminated by the lamp E, and was viewed through the optical aperture F with the aid of a telescope with a reading device, or was photographed by a camera with a chamfered plate.

In Fig. 3 is shown one of the photographs taken of sample No. 1, which was a single crystal of pure tin (resistance at helium temperatures $\rho_{4.2°K} = 9 \times 10^{-5} \rho_{20°C}$). Over most of the surface of the disc the structure is satisfactorily close to a system of parallel layers. In spite of the presence of a certain amount of branching, the average period of the structure, determined over a length of 20 — 25 mm in various portions of the disc, turns out to be constant to an accuracy of 3 — 4%.

The region of irregular structure, with a large percentage of the n-phase in the lower part of the surface, is separated from the remaining part of the picture by a clear boundary, which represents the projection of the edge of the reverse side of the disc, brought about by the magnetic field penetrating the sample at an angle.

A structure of simple form was obtained only in a definite range of variation of the relative magnetic field h ($h = H_0/H_c$, where H_0 is the external field). For $h < 0.91 - 0.92$ (for $T = 2.165°$) and for $h < 0.8 - 0.85$ (for $T = 3.2 - 3.5°$), a complex structure arose. In Fig. 4, for example, it is quite apparent that small inclusions of the s-space have developed inside the n-domains.

For fields approaching the critical field ($h > 0.99$), the laminar structure is gradually replaced by a system of s-filaments. In Fig. 5 is shown an intermediate stage, in which filaments and laminae occur together.

For a laminar structure of the type shown in Fig. 3, which has a definite period, the quantity Δ can be determined from the experimental data, since the results calculated by Landau[1] can be used also when there is an inclined magnetic field. It can easily be shown that the geometrical shape of the layers remains the same as for a perpendicular field (Fig. 1), but Eq. (1) becomes

$$\Delta = (a^2/L)\,\varphi(\eta)\,(1 - h^2\cos^2\beta), \tag{2}$$

where

$$\eta = a_n/a = h\sin\beta\,/\sqrt{1 - h^2\cos^2\beta}.$$

Thus, to determine Δ it is necessary to know not only a, L, and ρ, but also the quantity h, with possibly greater accuracy since the difference $1 - h^2\cos^2\beta$ is small in our case. Because of this, H_c was determined in each experiment upon the disappearance of the last traces of superconducting filaments (with an accuracy of approximately 0.2%).

The principal measurements were carried out with sample No. 2, a single tin crystal of greater purity than sample No. 1 ($\rho_{4.2°K} = 1.4 \times 10^{-5}\rho_{20°C}$). The crystalline axis of 4-fold order lay in the plane of the specimen and was directed along the projection of the magnetic field onto the plane of the sample. In Fig. 6 we plot the results of one series of measurements at T = 3.4°. With the aim of showing up the influence of non-equilibrium of the state on the results, different methods were used to carry the specimen over to the point of the intermediate state under study: the field was decreased at constant temperature (whereupon it was necessary for new layers to arise and for the period a to diminish), or the field was increased. In all cases laminar structures developed, aligned in the direction of the projection of the field. It was also found, however, that as a rule the values of a exhibited an hysteresis, which was more noticeable for the higher values of η. The actual value of Δ ought to lie in between the results obtained for these transition modes. Transitions achieved by varying the temperature at constant field gave results analogous to those for a changing field. We also tried to decrease the influence of hysteresis by causing a transition by varying the field about the required value (points marked with small circles); the field was first changed by about 1%, and then the amplitude of oscillation was gradually decreased to zero. The period of oscillation amounted to 30 — 60 seconds, which was sufficient for the structure to be able to follow the changes of the field.

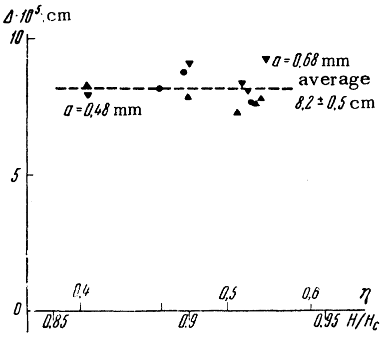

FIG. 6. Sample No. 2, T = 3.4° K. ▼ — decreasing field, ▲ — increasing field, ● — diminishing oscillating field.

It is difficult to make any definite statement concerning the origin of the hysteresis effects in our experiments. In every case chemical impurities, and in general those dislocations of the periodic lattice for which a strong effect shows up in the magnitude of the residual electrical resistance, apparently did not play an important role, since there was no improvement observed in going from sample No. 1 over to sample No. 2, which had a considerably smaller residual resistance. The surfaces of the samples were mirror smooth and probably free from strain, since the cast specimens separated quite easily from the mold. (The mold was made of glass, whose polished surface, next to the casting, was covered first with a film of oil a few molecular layers thick and then with a barely noticeable layer of carbon black.)

The absence of the requisite number of nucleii for one phase or the other cannot be invoked to explain the hysteresis, since on every picture there were observed several branches, i.e., nucleii virtually already formed.

Except for the hysteresis effects, the principal source of error in our measurements appears to be the fact that the specimen was not the infinite lamina to which Eq. (2) is strictly applicable. We can estimate the magnitude of this error by also carrying out a calculation in which we take the specimen to be an ellipsoid with an axis ratio of $1/25$. The data obtained for Δ are in this case smaller by 5 — 10% (in relation to the quantity h) when compared with the reduced values. However, since it is difficult to establish which method of calculation gives the most accurate results for the disc, we have not introduced a correction for the finite size of the sample, and have made use of Eq. (2). A detailed study of this question would make sense only if it were possible to make a considerable improvement in the reproducibility of the results.

In Fig. 7 are shown the results of measurements made at different temperatures. The uncertainty in the values of Δ can be estimated as about 6 — 8% on the side of increasing Δ, and 12 — 15% on the side of decreasing Δ. The results are most accurate at low temperatures. The asymmetry in the magnitude of the error is related to the fact that we have neglected to make the correction discussed above for the finite size of the disc. As is clear from the figure, within the limits of this error our results are described by the simple relation

$$\Delta = \frac{2.5 \cdot 10^{-5}}{\sqrt{1 - T/T_c}}. \qquad (3)$$

The results of measurements on sample No. 2 agree well with the preliminary data obtained with sample No. 1. It should be emphasized that Eq. (3) is an asymptotic expression, valid only near T_c, and cannot be extrapolated to the region of much lower temperatures.

We also carried out a series of measurements with specimen No. 2 at $T = 3.4°$ for different orientations of the axis of 4-fold order relative to the magnetic field (the sample could be turned about in its plane during an experiment). Within the limits of error, the results did not depend on the angle of deflection. It should be pointed out that these measurements were in the nature of a control experiment, and do not provide essential information concerning the anisotropy of Δ, since changes of the angle between the superconductivity current (directed near the axis of rotation) and the crystalline axes are negligible here.

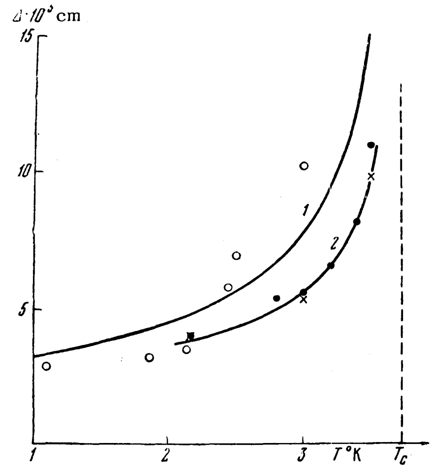

FIG. 7. ● — sample No. 2; × — sample No. 1. ○ — Shawlow's data. Solid curves: 1 — from theory of Ginzburg and Landau; 2 — from Eq. (3).

From the experimental data concerning Δ obtained recently by other authors, we have plotted in Fig. 7 the values obtained by Shawlow,[5] who studied the structure of the intermediate state with niobium powder. Shawlow's method has, however, a fundamental objection to it, and in our opinion his data are not sufficiently reliable. Shawlow ignored the appearance of a fine structure in the layers which he observed, apparently because the powder he used to bring out the pictures was too coarse. The specimens which he investigated were polycrystalline ones, with which appreciable hysteresis effects are associated, and one cannot estimate the magnitude of this hysteresis because placing the superconducting laminae perpendicular to the field, as Shawlow did, makes it impossible to obtain a simple well-ordered picture for the different transition modes. It turns out also to be impossible to use a single crystal in this case, as mentioned by the author himself.

A preliminary study of the structure of the intermediate state under various conditions[3] forced us eventually to reject Shawlow's method, which was, at first glance, the most natural one.

Certain information concerning the order of magnitude of Δ and its temperature dependence has been obtained by indirect methods. Shal'nikov,[6] the first to arrive at an estimate of the magnitude of Δ, used a bismuth probe to study the magnetic field in gaps of different widths formed between tin hemispheres. His estimate for Δ amounted to $0.6 - 1.5 \times 10^{-5}$ cm for $T = 3.1°$ K. However the theoretical assumptions concerning the degree of uniformity of the magnetic field over the width of the gap, on which his work was based, can nowadays be regarded as justified only qualitatively.

Desirant, Schoenberg, Andrew, and Lock[7,8] have determined the order of magnitude of Δ by analyzing the shape of the magnetization curves of small specimens. The indefiniteness of the theoretical premises concerning the structure of the intermediate state, and also, apparently, the marked effect of the lack of an equilibrium state in the sample, led to large dispersion of the data, which differed among themselves by as much as a factor of 3 or 4 or more. Most of the data obtained were still two times smaller than our data for the same temperature.

Faber[9] studied the dependence of Δ on temperature by measuring the rate of enlargement of nucleii of the superconducting phase in a supercooled specimen. The difficulty of carrying out this process, as the author noted, made it possible to estimate the absolute value of the magnitude of Δ only to within 50%, but the relative data concerning the temperature dependence appear to be more accurate (to within about 5%). These data are in good agreement with our measurements, since the ratio of our value of Δ to that given by Faber is equal to 1.65 over the entire temperature interval, with unsystematic deviations of less than 5% for individual points.

In Fig. 7 is also plotted a curve for Δ calculated according to the theory of Ginzburg and Landau[10] with account taken of the more accurate approximation to the relation between Δ and the penetration depth δ, obtained by Ginzburg.[11] The value of δ measured by Laurmann and Schoenberg[12] was used. The ratio of the theoretical value of Δ to the measured one is 1.5 near T_c and depends substantially on the temperature, decreasing to 1.2 at 2.165° K. This difference, although certainly not large, is nevertheless outside the limits for the probable experimental error for the measurements of Δ and δ (about 5 – 8% in the experiment of Laurmann and Shoenberg[12]). It should be noted that other measurements by the author[13] have given a value of δ approximately 8% smaller, and the recent work of Chambers[14] a value 16% smaller than that of Laurmann and Shoenberg.[12] Such a decrease in the value of δ would lead to a further increase of the discrepancy between theory and experiment, since Δ is approximately proportional to $1/\delta$.

The possible explanation of the disagreement as a result of the anisotropy of the superconductive properties of tin is not a very likely one, since the anisotropy of Δ according to preliminary data does not exceed 10%.[15] The anisotropy of δ in the estimates which have been made also is not sufficiently large. Nonetheless, the problem of the nature and magnitude of the anisotropy of Δ and δ is at present completely confused, and its solution is necessary for clarifying the source of the observed discrepancy and for directing further improvements of the theory.

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