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Structure of the Intermediate State of Superconductors

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The structure of the intermediate state was studied by application of fine ferromagnetic powder to the surface of a tin specimen. Two-dimensional pictures of structures of various types were obtained for various contents of normal phase in the specimen. The influence of a number of other factors (method of transition, temperature, specimen size and others) on the character of the picture was also studied.

INTRODUCTION

As is well known, a superconductor in the intermediate state is broken up into regions or "domains" of superconducting and normal phases (s – and n – phases). The magnetic field is concentrated almost entirely in the n – domains. Thus the field outside the specimen and very close to its surface is also nonuniform. The dimensions of the s – and n –domains in thermodynamic equilibrium must be determined by two basic factors which act in opposing directions. The field energy close to the specimen will be reduced by reduction of thefield inhomogeniety and from this point of view it is energetically favorable to increase the degree of disperseness of the state. On the other hand the presence of a positive surface tension at the phase boundaries tends to increase the dimensions of the domains.

Agreement between experimental and theoretical investigation of the geometry of the intermediate state would therefore permit the development of a method of finding the magnitude of the surface tension from observation of the equilibrium dimensions of the structure. A knowledge of this quantity and its dependence on various factors (temperature, crystallographic orientation of the phase boundary and others)would be very useful for the development of the theory of superconductivity.

This problem however is a long way from being solved. Mathematical difficulties prevent consideration in a general form of the problem of the shape of the phase boundaries. Landau succeeded, however, in giving exact calculation of the shapes and sizes of the domains under certain simplifying conditions. In one calculation nearly plane parellel s – and n – layers were considered,[1] while in another[2] a model was considered in which the n – layers repeacedly branched as they approached the external surface so that the structure was homogeneous or "mixed" at the surface. Subsequent work by other authors[3,4,5,6] is based on the methods used by Landau. In particular it was shown[4] that a model with layers branching a limited number of times and coming out to the surface without formation of the mixed phase (i.e., in a certain sense a combination of the models of Refs. 1 and 2) is energetically more favorable than the original methods. In one way or another all the formulas proposed for connecting the surface tension with the dimensions of the domains, have been obtained only under various simplifying assumptions, still requiring experimental verification, about the shapes of the domains.

The experiments of Shal'nikov, Meshkovskii and Tumanov[7-11] on the pattern of the field distribution in a gap between two tin hemispheres by the methods of a bismuth micro-probe and ferromagnetic powder have shown that the real structures of the intermediate state have not a regular or simple form and are not reproducible for various modes of transition to the given point in the intermediade state (for various procedures of changing the field and temperature). The $s-$ and $n-$ domains in these experiments have a range of sizes down to very small sizes. From other considerations Meshkovskii deduced the existence of a "fine structure" within the $n-$ layers observed by him (which were of width of order 1 mm and predominantly radial in direction), i.e., the existence of a large number of fine unresolved inclusions of $s-$ phase. The size of the domains was not determined in these experiments both because of the irregularity of the patterns and because of insufficient resolving power of the methods used. It should be noted that in spite of the irregularity of the general pattern of distribution of the domains, an averaged characteristic of the state such as the relative content of normal phase in the specimen (denoted in what follows by η) is practically a single valued function of H and T (at least for pure macroscopic spherical specimens of "soft" superconductors, such as tin). It was also shown[12] that the average domain size in a monocrystalline tin specimen is well reproducible for different modes of transition to the given point in the intermediate state.

In the present work, we investigated the topography of the $s-$ and $n-$ domains at the surface of tin specimens for various values of η . Our main aim was to sort out which qualitative features of the real structure were associated with an equilibrium and stable character, having in mind a possible comparison of the results with the above-mentioned theoretical studies, where only the case of thermodynamic equilibrium was considered.

METHOD OF OBSERVATION

The form of the $n-$ domains was investigated by depositing a fine ferromagnetic powder on the surface of the specimen. Nickel powder, consisting of rounded particles of average size about 1 μ was deposited by sprinkling on the open surface during the actual experiment. In order to do this a glass tube B was introduced into the cap of the liquid helium Dewar (Fig. 1), and connected to it was a coarse-pored filter F into which some well dried nickel powder was sprinkled.

The experiment was carried out as follows. The specimen A was brought into the desired point of the intermediate state by suitable variation of field and temperature and then by means of the pump H helium was blown through the filter F ; some of the smallest particles of the powder were then carried by the current of helium into the Dewar and on to the surface of A . Observation

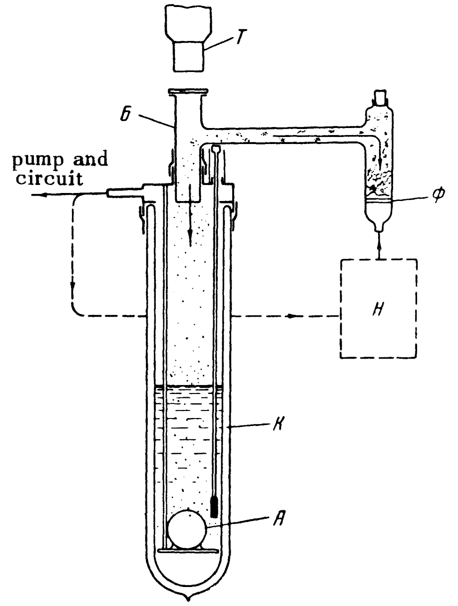

FIG. 1. Scheme of apparatus.

and photography of the patterns produced was carried out through the telescope T . In order that boiling of the helium should not disturb the photography, the pressure in the Dewar was increased during the time of exposure. After exposure, the powder could be removed from the surface by means of the brush K, and the experiment could then be repeated. Before each experiment the specimen was heated above the transition temperature. The powder clung rather firmly to the specimen surface and this made it possible to photograph patterns even after the specimen was taken out of the apparatus.

The resolving power of the method was checked by means of a special model prepared from alternating layers of copper and lead of various thicknesses in the range from 1 to 0.05 mm, and put into the apparatus in place of the specimen. The pattern obtained at 4.2 °K in a field of 100 Oe reproduced the structure of the model very well.

For studying the distribution of domains in the gap between two hemispheres we used a method of Shal'nikov[9,11]. Nickel powder was introduced into the gap before the experiment and during the experiment it redistributed itself when the specimen was shaken, being drawn into the regions of maximum field.

RESULTS

The structure of the intermediate state observed in our experiments proved to be very complicated and varied, and it is possible here only to describe the most clear-cut qualitative characteristics of these structures.

Figures 2 to 4 show the patterns obtained on the surface of a monocrystalline tin sphere of diameter 30 mm and of purity 99.998% for $\eta = 0.15$. It can be seen that the n-domains issue from the surface in the form of narrow wrinkled strips. For still smaller η, slose to 0, the n-domains are scattered over the surface in the form of separate small regions of similar wrinkled strips.

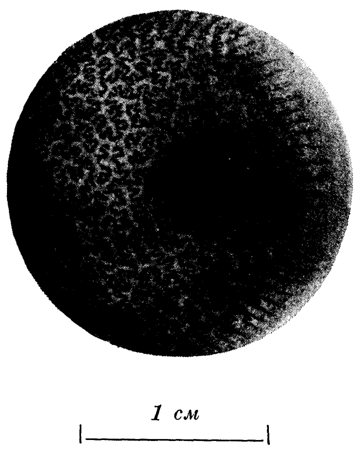

FIG. 2. General view of sphere of diameter 30 mm for $\eta = 0.15$, $T = 2.85^\circ$ K, Transition $n \to s$. H = const.

FIG. 3. Part of Fig. 2, greatly enlarged.

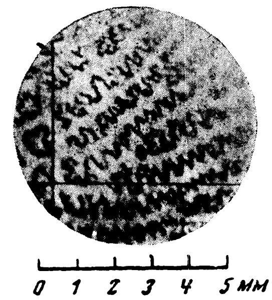

FIG. 4. Part of the surface of a sphere of diameter 30 mm close to a pole; photographed through the telescope; $\eta = 0.15$, $T = 2.85^\circ$ K, Transition $s \to n$, T = const.

With increase of η the wrinkled n-domains cover an even larger fraction of the specimen surface and their form gradually changes in such a way that completely closed little islands of s-phase are formed between them (Fig. 5). The type of structure for η close to 1 is shown in Fig. 6. No fine structure in the n-phase surrounding the rounded islands of s-phase visible in the picture is observed, and does not apparently exist. An indirect proof of this comes from a rough estimate of the relative area occupied by the n-phase in the photograph, and this agrees within an accuracy of 10% with the value of η determined from the diagram of state.

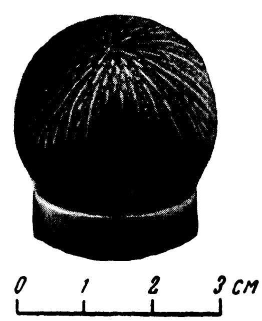

FIG. 5. General view of sphere of diameter 30 mm for $\eta = 0.72$, $T = 2.85^\circ$ K, Transition $s \to n$, T = const.

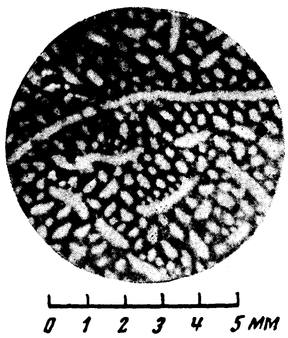

FIG. 6. Part of the surface of a sphere of diameter 30 mm close to a pole, photographed through the telescope; $\eta = 0.8$, $T = 2.85°$ K, Transition $n \to s$, H=const.

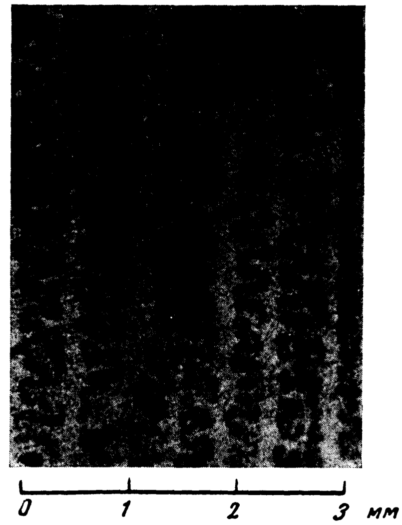

FIG. 7. Pattern in a gap of width 0.3 mm between hemispheres of diameter 40 mm. $\eta = 0.145$, $T = 2.36°$ K, Transition $n \to s$, T = const.

These qualitative characteristics (wrinkling of the n-domains for small η and "island-like" s-domains for η approaching 1) were observed whatever the mode of transition to the intermediate state, on monocrystalline specimens with electropolished or etched surfaces and on polycrystalline specimens with mechanically polished surfaces. In the latter case, however, the domains were somewhat more markedly wrinkled for small η, while for $\eta \sim 1$ they were somewhat less rounded than for the monocrystalline specimens.

We also varied the size and shape of the specimens. We investigated tin spheres of diameter 40, 30 and 12.6 mm diameter, and tin toruses and cylinders of diameters from 10 to 4 mm in transverse fields. Experiments were also made with a lead sphere of 40 mm. The specimen temperature was varied from 2.2 to 3.4° K.

In all these experiments the above-mentioned features were observed, and this supports the view that these features are characteristic of the equilibrium structure of the intermediate state.

In order to study the distribution of domains inside a superconductor we made experiments in the gap between two monocrystalline tin hemispheres of diameter 40 mm and of purity 99.9%. Gaps of width 0.3 and 0.1 mm gave no new qualitative results as compared with the external surface, both for large and small η (Fig. 7).

There is every reason for supposing that the wrinkled structure of the domains observed at the surface for small η is not preserved in the interior of the specimen, but that the domains tend to thicken, thus reducing the total surface area of the boundaries between s- and n-phases and consequently, also reducing the free energy. Thus the n-layer must have the form schematically illustrated in Fig. 8. In this connection a gap of 0.1 mm for a sphere of diameter 40 mm must be considered as insufficiently narrow for properly showing the character of the distribution within the specimen, i.e., the question of the critical gap width raised in Refs. 2 and 7 is once again relevant. In one experiment with a very narrow gap (0.05 mm) we did succeed in getting a pattern of nonwrinkled, comparatively broad layers (not sufficiently perfect, however, for photographic reproduction), and this apparently confirms the hypothesis proposed. The reason for the occurrence of wrinkling of domains at the surface, must presumably be analogous to the reason for the branching of the layers as they approach the surface in the model[4]. In both cases the disperseness of the structure grows towards the surface, and the field close to the surface becomes more uniform, which is favorable because of the accompanying reduction of the free energy of the state.

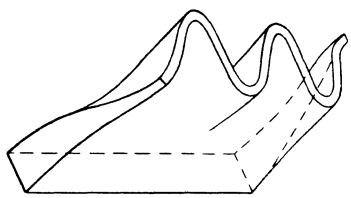

FIG. 8. Proposed form of an n-layer for small η.

The patterns obtained for various modes of transition to the given point of the intermediate state have besides the above-mentioned common features also certain marked differences. We have tried the following modes of transition: (a) cooling in a constant field ($n \to s$, H = const), (b) increase of field at constant temperature ($s \to n$, T = const); and (c) reduction of field at constant temperature ($n \to s$, T= const).

The influence of the mode of transition shows itself appreciably only in the general disposition and form of the coarser details of the structure (regions of fine structure as a whole and large s-domains), which for transition of type (b) were disposed in meridional directions over the whole surface right up to the "magnetic poles" (see Figs. 4 and 5). An analogous pattern, i.e., radial disposition of the domains, was observed in the gap (see Fig. 7). For the cylinders (and toruses) in transverse fields the coarse details of the structure were arranged perpendicular to the cylinder axis. For transitions of types (a) and (c), no such oriented distribution of the domains was observed (see, for instance, Figs. 2, 3 and 6).

It may be supposed that this difference is connected with the fact that in one case the transition starts from the completely superconducting state and in the other from the normal state. The general picture is apparently determined by the kinetics of the transition and the conditions of growth of the s- and n-domains. It is possible that the character of the patterns is influenced by the orienting action of the currents induced in the specimen at the moment of transition*. The clarification of the influences of these factors demands, however, a more detailed investigation.

Over a small part of the surface of the sphere close to its "magnetic equator" (in the lower part of the photographs of the sphere in Figs. 2 and 5) the pattern has a character different from that over the remaining larger part of the surface. Here the n-domains, independently of the mode of transition, always have the form of relatively broad unwrinkled strips of a meridional direction. Similar observations were made on the toruses and cylinders. Apparently this form of the layers at the equator is a peculiarity characteristic of the equilibrium state and connected with the smallness of the angle between the surface of the specimen and the magnetic field at these points. As was shown by Dzialoshinskii[6] for layer-like structures of the type of Ref. 1, in the case of a plate at a small angle to the field, the planes of the layers must be perpendicular to the external surface. For a sphere this condition is evidently satisfied by a meridional disposition of the layers. The thickness of the layers must grow at small angles, since in this case the inhomogeneity of the external field, connected with the existence of layers must also diminish (for plates or cylinders parallel to the field, the inhomogeneity evidently disappears altogether). Thus in this case the factor leading to wrinkling of the layers is weakened.

CONCLUSION

From our observations the conclusion can be drawn that the real structure of the intermediate state is considerably more complicated and varied than that of the theoretical models constructed up to now. This comes about not only because the structures achieved in practice are insufficiently completely equilibrium ones, but mainly because the equilibrium structures with least free energy are really highly complicated and varied in type for various forms of specimen and various η.

The theoretical calculation of the structure for all values of η would seem to be a very difficult problem as is the problem of quantitative treatment of the experimental results in the general case. The case of a specimen whose surface makes a small angle with the field, and the cases $\eta \to 1$ and $\eta \to 0$ are relatively much simpler and more suitable for quantitative investigation.

Undoubtedly other possibilities also exist for investigating the intermediate state apart from the direct observation of the shapes and sizes of the s- and n-domains (see, in particular, Refs. 13, 14 and 15). It should be noted, however, that in these cases also, an exact quantitative interpretation of the results requires a knowledge of the geometry of the structure of the intermediate state.

* We compared the patterns obtained on a torus and on the same specimen after transformation into a "horseshoe" after cutting a gap in it. In the second case, with other conditions remaining the same, the pattern seemed less ordered.

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