Search for Surface Superconductivity Induced by an Electric Field*

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Using the electrical and thermal conductivity of rather thick films of tin, the possibility of surface superconductivity induced by the presence of an electric field normal to the surface of the film has been investigated. No such effect was found.

I. INTRODUCTION

THE existence of surface superconductivity has first been predicted by Saint James and DeGennes[1] and subsequently been verified in numerous investigations.[2–8] If the Ginzburg-Landau[9] constant κ is larger than 0.417, then a superconducting surface layer is stable in a magnetic field strong enough to quench superconductivity in the interior provided that the superconductor bounds on vacuum and not on a normal metal. The thickness of these surface layers is about twice the coherence distance ξ_0 or about 1800 Å for lead[6] (Pb). If tin were to exhibit surface superconductivity one would expect this surface layer to be about 4600 Å thick. Another type of surface superconductivity has been proposed by Ginzburg.[10] He proposes that a dielectric layer (e.g., an oxide layer) on the surface may induce surface superconductivity. Strongin *et al.*[11] have found some experimental evidence for the actual occurrence of this effect.

Walther Meissner[12] investigated the effect of an electric field (normal to the surface) on the transition of a rod of tin in a magnetic field. Of his numerous observations, only the following will be considered here. He measured the change of flux $\Delta\Phi$ as a function of temperature in a search coil surrounding the sample when the (longitudinal) magnetic field was changed by an amount ΔH. When no electric field was applied he found the usual hysteresis.[13] Upon application of an electric field the hysteresis disappeared completely. The flux penetration occurred always at the temperature where formerly the transition was found for decreasing temperature. The effect was independent of the direction of the electric field.[14]

One possible explanation for this observation is the assumption of Saint James–DeGennes–type surface superconductivity induced by the electric field. A superconducting surface layer undergoes at the upper critical field H_{c3} a second-order phase transition which precludes any hysteresis. Theoretical considerations also lead in the same direction. W. Meissner uses a simple free-electron model to explain his observations. His model is somewhat reminiscent of the "third Ginzburg-Landau equation" as derived by Anderson, Werthamer, and Luttinger.[15] These authors have shown that a gradient in the chemical potential (that is in the number density of the superconducting electrons) results in an electrical potential. An externally applied potential results in a concentration of electrons at the surface and an internal potential gradient over the distance of a Debye length, and possibly surface superconductivity.

As was shown in Ref. 6 (hereafter referred to as S.M.) the simultaneous measurement of the electrical and thermal conductivity of thin-film samples is ideally suited for the study of surface superconductivity and is used here to investigate whether surface superconductivity occurs in films of tin if an electric field is applied to one surface.

II. EXPERIMENTAL ARRANGEMENT

The arrangement is almost identical to that used by S. M. Two samples, one for the measurement of the electrical, the other for the measurement of the thermal conductivity are mounted on a copper rod extending from an inner helium vessel into the vacuum jacket surrounding it. Both samples are films of tin simultaneously vacuum-deposited onto microscope cover

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[14] This is somewhat astonishing in view of the work of Glover and Sherrill [Phys. Rev. Letters **5**, 248 (1960)] who observed an increase of the critical temperature T_c of very thin films of tin for negative charging, and a decrease of T_c for positive charging. Similarly, one would expect that negative charging would favor the formation of surface superconductivity, but that positive charging would suppress it. A change of T_c or H_c through charging is not expected for the bulk samples used by W. Meissner and is also not expected for the rather thick films used here.
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glasses. A gold film was similarly deposited onto the other side of the glass. During evaporation of the metals, the substrates were cooled to liquid-nitrogen temperature. A liquid-helium-filled trap kept the pressure during the evaporation of the tin below 1×10^{-6} Torr. The "electrical" samples had a width of 1.1 mm and a length of 8 mm between potential taps. In order to obtain sharp transitions the penumbra was trimmed off. The "thermal" samples were provided with two carbon thermometers, one serving as heater-thermometer, the other as reference thermometer. They were connected in a Wheatstone bridge arrangement.

The "tail" of the vacuum jacket (which contained the copper rod with the samples) extended into a precision wound niobium coil which provided a longitudinal magnetic field, parallel to the film and to the flow of electrical or heat current.

The temperature of the inner helium bath was electronically stabilized and was constant to a small fraction of a millidegree. The electrical transitions were recorded by plotting the potential accross the sample vs the magnetic field on an x-y recorder. For each temperature 3 recordings were taken: one without any potential applied between the tin and the gold backing and one each with the gold backing at a potential of $+75$ or -75 V relative to the tin.

Two series of runs were made, the first with samples of 3830 Å thickness the second one with samples of 6455 Å as determined by interferometry of equal chromatic order. Figure 1 shows typical recordings of the transitions of the electrical samples for the 6455 Å film.

When an attempt was made to record the thermal transitions similarly by plotting the error signal of the Wheatstone bridge (balanced at zero field) as a function of the magnetic field, rather poor traces were obtained because of flux motion in the film. Point-by-point measurements gave clearer results and are shown in Fig. 2. Again three transitions at zero, $+75$- and -75-V potential between the tin and the gold are shown.

The continuity of the connections to the gold and tin films was checked by measuring the capacitances of the various combinations during the run.

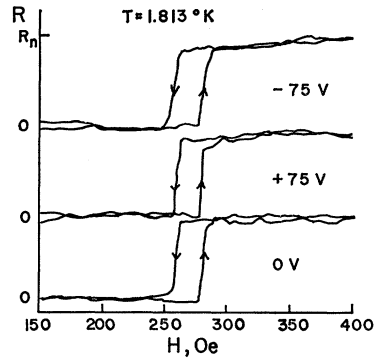

FIG. 1. Resistance of a 6455 Å film of tin as a function of the magnetic field with the gold electrode facing the tin at a potential as indicated on the curves.

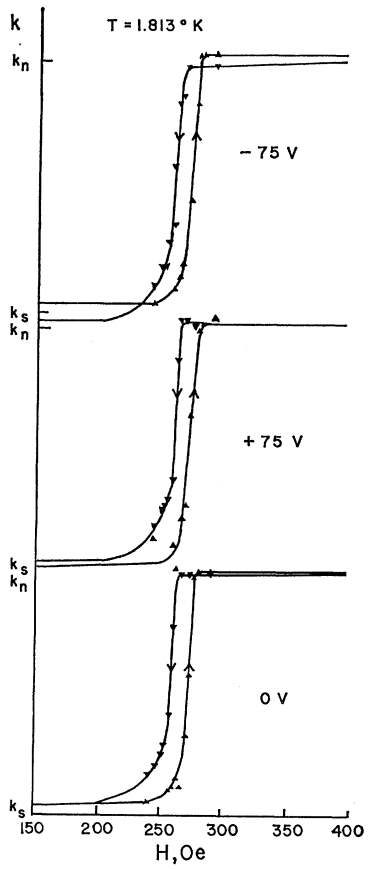

FIG. 2. Thermal conductivity k of 6455 Å film of tin as a function of the magnetic field with the gold electrode facing the tin at a potential as indicated on the curves.

The rather low voltages were chosen in conformity with those used by W. Meissner. The thickness of the cover glass was 0.12 mm for the thermal samples and 0.15 mm for the electrical samples. Meissner used 65 V and (as evaluated from the capacitance between sample and facing electrode) a distance between sample and electrode of 0.133 mm.

As can be seen from Fig. 1 and 2 the transitions are unaffected by the presence of the electric field. The hysteresis is present in all cases. The current of 12 μA used for the measurements of the electrical transitions is small enough not to destroy surface superconductivity.

III. DISCUSSION

The results here show that the absence of a hysteresis observed under the influence of an electric field does not occur with all experimental arrangements. It is also possible that the difference results from the use of a thin film rather than a bulk sample. The quality of the samples used here should have been as good as that of the polycrystalline sample used by W. Meissner. The present samples were quite shiny and smooth. The residual resistance ratio of the 6455 Å sample was $R_{4.2}/R_{273}=4.37\times10^{-2}$, part of which is accounted for by boundary scattering. The critical temperature of

3.832°K indicates that the sample is somewhat under stress. The width of the transition was 0.028°K. While the thickness of the first set of samples (3830 Å) was less than $2\xi_0=4690$ Å (for tin) the second set of samples was sufficiently thick (6455 Å) so that on the side with the electric field a superconducting layer could have been formed with some of the tin still remaining normal-conducting. If this had happened, it would have led to observations quite similar to those of S.M. If the surface superconducting layer had been much thinner, the first set of samples would have given better accuracy. In neither case was any sign of surface superconductivity detected.

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Superconductivity in the Alloy System Indium-Thallium*

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The superconducting transition temperatures of over 75 samples spanning the entire composition range of the indium-thallium alloy system have been measured. The highest T_c measured was about 3.8°K in the bcc phase; the lowest was 2.5°K in the fcc phase. In all phases, T_c decreases with increasing Tl content, the rate of decrease in the tetragonal (indium) phase being, however, much less steep than in the other phases. The Seraphim empirical rule, $\Delta T_c=k_1x+k_2\ln x$, is found not to describe our data in the dilute thallium range. An anomaly in the T_c-versus-composition plot at 4.4 at.% Tl is extremely similar in form to one previously identified in In-Cd as a Brillouin-zone effect. Transition temperature and isothermal magnetization data are used to locate the two-phase region separating the tetragonal and cubic phases; the boundaries of this region are: 30.1±0.4 and 31.7±0.3 at.% Tl at 3.3°K. The existence of such a region indicates that the tetragonal-cubic transformation is first-order, in agreement with the earlier room-temperature calorimetric work of Predel. The transition temperature of bcc Tl was estimated by extrapolation of the bcc alloy data to zero solute concentration; the result was 2.49°K.

I. INTRODUCTION

THE alloy system indium-thallium has been the subject of many investigations. This system is popular with physicists and metallurgists interested in the electronic structure of metallic alloys for several reasons. The important reasons are: (1) both In and Tl are reasonably "simple" metals, i.e., their conduction-electron dynamics approach those of free electrons, and (2) they are nomially isoelectronic, each having three electrons per atom available for metallic conduction. Thus, the alloys all have three electrons/atom, independent of composition. The extent to which this is meaningful is not clear, but it certainly is true that thallium as a solute in indium causes a smaller change in almost any physical property than any other solute metal. The system is also convenient to handle experimentally, the metals are available in high purity, solid solubilities are extensive, temperatures are in a convenient range, etc. Not many properties, however, have been measured over the entire composition range except for the basic constitution diagram information,[1] the electrical resistance,[2] and the superconducting transition temperature (T_c).[2]

We undertook this work as one of a series[3–8] of studies of the electronic structure of nontransition metal alloys through measurement of the superconducting transition temperature. This technique in other alloy systems has lead to the discovery of electronic structure effects, e.g., Brillouin zone overlaps[9] and an order-disorder transformation.[10] It has also proved useful in investigating

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