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Paramagnetism of Tin Observed at the Superconducting Transition*

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Synopsis

Measurements have been made of the paramagnetism of tin cylinders in the presence of an external magnetic field and with an externally supplied current at the superconducting transition. It has been ascertained that the paramagnetic effect is not such an apparent one as once supposed, but an intrinsic one without hysteresis. The current minimum I_0, required for the appearance of a paramagnetic effect is represented in the $(I–H–T)$ space by the simultaneous equations $I_0=\xi\gamma d(T_c-T)$ and $H_0=\xi(T_c-T)-I_g/\gamma d$. Here I_g, γ, T_c and ξ are characteristic constants of the superconductor and have values 1.2 amp, 0.23, 3.73°K and 1.1×10^2 oersted/deg respectively for the case of tin. H_0 and d are the external magnetic field in cersted and the specimen diameter in mm respectively. It is shown that the formula, $I_0=I_g+\gamma dH$ obtained by earlier investigators for the minimum current requirement is the one for the orthogonal projection on the $(I–H)$ plane of the critical line in the $(I–H–T)$ space. The paramagnetic region is shown schematically in the $(I–H–T)$ space. Finally some remarks concerning the theory of the paramagnetic effect are given.

I. Introduction

Two important characteristics of the superconductor are the disappearance of its electrical resistance and of its magnetic induction below the critical temperature. Onnes came to the conclusion that the resistance of mercury vanished below 4.15°K, by measuring the potential drop between two points of the specimen and the current flowing along it. Afterwards Meissner and Ochsenfeld discovered that the superconductivity was accompanied by zero induction in the magnetic field below the critical field H_c, appropriate to the temperature. The independent variables of the state which may determine whether a metal is in the superconducting state, if it becomes one at all, are the temperature T, the magnetic field H, and finally because a current in the specimen may produce its own field H_I, we must consider the current I as a variable. Measurements of the resistance of a superconductor in a magnetic field whose direction was along the specimen cylinder was carried out by Alekseyevsky(1).

Experiments on the magnetization of a superonductor along which a current flowed was performed by Steiner and Schoeneck(2), who first observed the increase of magnetic induction in the specimen immediately before the superconducting transition. It is quite natural to expect that a superconductor, along which an

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A short report on the subject appeared in Phys. Rev., **98** (1955), 938.

(1) N. E. Alekseyevsky, J. Exp. Theor. Phys. U. S. S. R., **8** (1938), 342.

(2) K. Steiner and H. Schoeneck, Phys. Z., **44** (1943), 346.

electric current flows in the presence of an external magnetic field, may become normalconducting in the external field smaller than H_c, because the effective magnetic field which destroyes the superconductivity may be the resultant of the external magnetic field and the magnetic field due to the current. It was, however, quite unexpected that a superconductor showed a large paramagnetic susceptibility(3) preceding the change to diamagnetism. Indeed, Steiner and Schoeneck observed the so-called paramagnetic effect with a cylindrical superconductor along which a current larger than a critical one flowed in the presence of an external, longitudinal field. Afterwards Steiner(4) reported that there was a relation for the occurrence of the paramagnetic effect between the current and the external field and that he observed the same effect also with a hollow cylindrical superconductor along which a current larger than a critical one flowed in the presence of an external, circular magnetic field perpendicular to the specimen axis.

It was, however, conceived once(5) that the paramagnetic effect might have possibly been only an apparent one, because in earlier works the observation of the paramagnetic was coupled with a simultaneous variation of one such as H or T of the variables of the state. It was hoped to investigate the actual existence of the paramagnetic effect without resorting to a simultaneous variation of one of the variables.

Meissner et al.,(6)(7) who retraced the work of Steiner and Schoeneck first by measuring the magnetization of the specimen in the magnetic field which was reversed at fixed values of temperature and current, emphasized that the paramagnetic effect was not an apparent but an intrinsic one from their subsequent measurements by the fluxmetric recording method in which the temperature was changed very slowly at fixed values of current and field, because they confirmed that the flux increase could be maintained permanently if current, field and temperature were kept constant. Furthermore they clarified that the coefficient of the magnetic field appeared in the formula for the minimum current requirement proposed by Steiner was proportional to the specimen diameter. They performed measurements on hollow cylindrical specimens(7), besides on cylindrical one, with the result that they could observe also the flux increase with both a search coil to measure the flux inside the hole and another one to measure the total flux through the specimen, and hence concluded that the flux increase was due not to the volume magnetization but to the circular component of current in the specimen. In order to verify this conclusion they performed experiments on both a cylinder and a hollow cylinder splitted so as to hinder the circular current, without

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(3) Tin is a paramagnetic metal at room temperatures. Its mass susceptibility is, however, of the small order of 10^{-8} and hence this paramagnetism can be discarded here. The paramagnetism discussed in this paper is of the same order as the absolute value of the perfect diamagnetism, the volume susceptibility of which is $-1/4\pi$.

(4) K. Steiner, Z. Natforsch., **4a** (1949), 271.

(5) For instance, K. Mendelssohn, Repts. Prog. Phys., **10** (1946), 358.

(6) Meissner, Schmeissner and Meissner, Z. f. Phys., **130** (1951), 521, 529.

(7) Meissner, Schmeissner and Meissner, Z. f. Phys., **132** (1952), 529.

observing the paramagnetic effect.

Thus all these earlier workers did not carry out measurements of the magnetization of a superconductor at fixed values of current, magnetic field and temperature, but did by changing one of three variables of the state at fixed values of remaining two. Although the fluxmetric recording made by Meissner et al. showed that the paramagnetic effect was not a dynamical one, it is quite desirable to investigate the effect without resorting to the method of changing any one of three variables. The confirmation of the effect by such a method may be, in conjunction with the confirmation by the fluxmetric recording made by Meissner et al., enough to disprove the question that the paramagnetic effect may be an apparent one probably due to the dynamical method of measurements. The formula for the minimum current requirement proposed by Steiner and extended by Meissner et al., as described above, is

$$I_0 = I_g + \gamma dH. \qquad (1)$$

Here I_g and γ, which are characteristic constants of the superconductor, are 1.2 amp and 0.17 respectively for tin, provided the specimen diameter d and the magnetic field H are measured in mm and in oersted respectively. According to this formula it seems always possible, irrespective of the temperature, to observe the paramagnetism with a current larger than I_0 for a fixed value of H. Actually, however, there is a one to one correspondence between H in *Eq.* (1) and the temperature T. Therefore the current minimum required for the paramagnetism should be determined as a curve in the three-dimensional (I-H-T) space, instead of by *Eq.* (1). Furthermore it seems there is no convincing measurement on the question of the existence of hysteresis phenomena in the effect.

The present investigation was undertaken in order to investigate whether the paramagnetie effect was such an apparent one as once supposed or not, and in particular the question of the existence of hysteresis phenomena in the effect, and to investigate the current minimum required for the appearance of the effect in the (I-H-T) space, which might serve as clues for the theoretical explanation for the effect. At present we have not yet a satisfactory theory of the paramagnetic effect. Finally some remarks on the qualitative idea and the formulated theory concerning the paramagnetic effect hitherto proposed are given, and it is shown that essentially new concepts seems necessary for the establishment of a satisfactory theory.

II. Experimental details

The experimental apparatus used in the present investsgations is shown schematically in Fig. 1. The magnetization of the specimens was measured with the use of a ballistic-type galvanometer in the same way as done in the case of ferromagnetic substances. We used following two procedures in accordance with our aim what a physical nature in the paramagnetic effect to study. A tin specimen S and a lead rod Pb(8) of the same diameter and length soldered together with Wood's metal at the upper end of S were placed in a uniform, external, longitudinal, magnetic field, and an electric current flowed down or up along them.

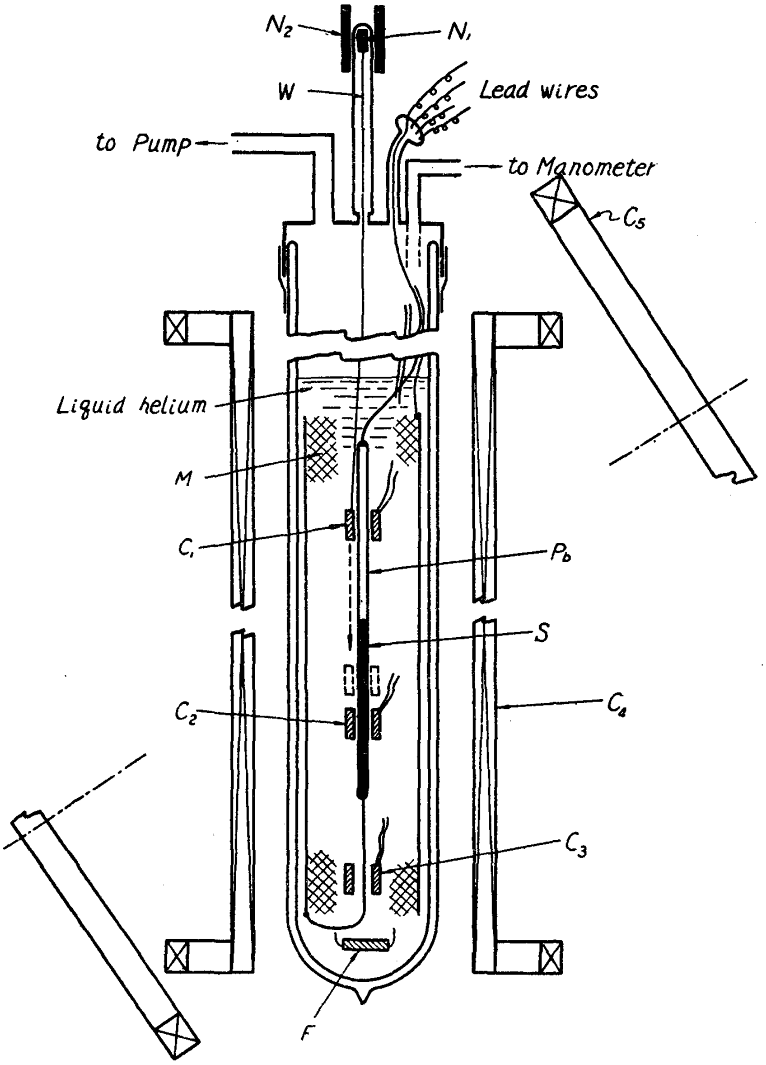

Fig. 1. Schematic diagram of the experimental apparatus. The outer Dewar vessel for containing liquid nitrogen is not shown.

We employed first the statical method, similar to that used by Mendelssohn et al.(9), of dropping a search coil in a uniform field from its position around the lead cylinder to a position around the tin cylinder. The difference in magnetic flux induced a current in a ballistic-type galvanometer which had a period of 3.6 sec, a resistance of 12.8 ohms and a critical damping resistance of 4.2 ohms. For the study of the intrinsic nature of the paramagnetism, measurements were carried out in such a way that two variables, the longitudinal magnetic field H and the temperature T, were held constant throughout, while the third variable, the current I through the specimen was changed in small steps. A movable search

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(8) In order to reduce the heat conducted through the current lead from outside the liquid helium Dewar, we joined S to Pb which has a small thermal conductivity, because its critical temperature is 7.26°K and it is in the superconducting state in the extent of I and H in which the present measurements were performed. Strictly speaking, the statical method described below, would give a relative paramagnetism of tin to lead, if the latter were paramagnetic in the region of I, H and T studied. It was, however, reasonably supposed that owing to the fact that the threshold field H_c for lead at temperatures studied is about 600 oersted, a large amount of current than used in the present measurements would be necessary in order to observe the paramagnetism in lead in the extent of magnetic field and at the temperature range concerned. Therefore it was safely assumed that lead was perfectly diamagnetic in extent of I and H in which the present measurements were carried out.

(9) Mendelssohn, Squire and Teasdale, Phys. Rev., **87** (1952), 589.

coil C_1 was dropped at each step. The coil C_1 was wound with 5,300 turns of BS#42 enamelled wire on a glass form. Nylon thread W was used to link the coil C_1 with a small N.K.S. magnet N_1 which could be moved with a hollow cylindrical N.K.S. magnet N_2 surrounding the gas-tight glass envelope housing the magnet N_1. The hysteresis measurement consisted in holding I and T constant throughout and reducing H in small steps from the value above H_c to zero, followed by a similar increase in H. The coil C_1 was dropped also at each step.

The second method i.e. the so-to-speak dynamical method used to determine the current minimum required for the paramagnetism consisted in holding T constant throughout, taking I as a parameter, and reversing H; the latter was changed in small steps. In this determination a search coil C_2 was fixed around the centre of the specimen, and a compensating coil C_3 connected in opposition to the search coil C_2 was fixed around a copper lead at a position sufficiently apart from the specimen in the uniform field. In this case also the difference in magnetic flux through two coils C_2 and C_3 induced a current in the ballistic-type galvanometer, when the magnetic field was reversed. The construction of C_2 and C_3 is quite similar to that of C_1.

The cylindrical copper mesh M was used as the return lead of the current in order to cancel the magnetic field due to the return current. A wire-wound solenoid system C_4 produced a uniform, longitudinal magnetic field which was homogeneous to within 0.07 per cent in the volume of the diameter 2 cm and the length 20 cm in the central portion of the solenoid system. The earth magnetic field was cancelled with a large Helmholtz coil C_5(10). A small heater F was placed in the bottom of the helium Dewar insuring thermal equilibrium throughout the liquid helium bath. A manostat(11) shown in Fig. 2 was used to fix the helium vapour pressure to within 0.2 mmHg. The pressure was measured with a mercury manometer and the temperature was determined by vapour pressure thermometry using the 1949 Mond Laboratory tables.

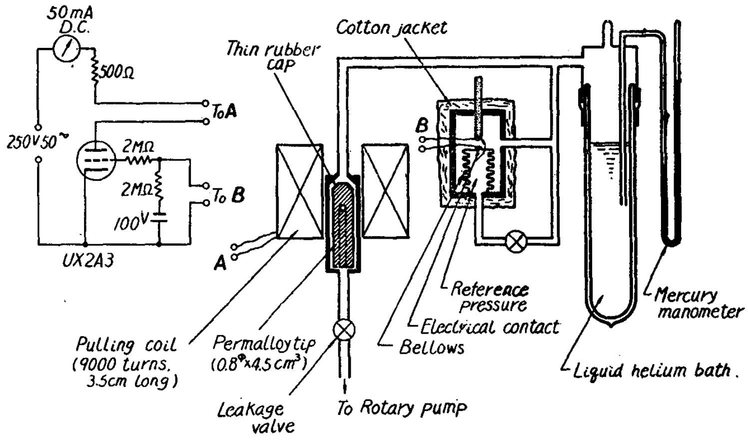

Fig. 2. Schematic diagram of the manostat used to fix the heluim vapour pressure to within 0.2mmHg.

The tin specimens were prepared from Johnson-Matthey spectroscopically pure tin (>99.995 per cent Sn). They were cast in vacuo in glass tubes of required

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(10) We feel grateful to Professor Y. Tanabe of the Research Institute for Scientific Measurements, Tohoku Univ., who gave us kindly the facilities of using the Helmholtz coil.

(11) We are indebted to Mr. K. Yasuhara, now at the Sumitomo Metal Works, Osaka, for the design and test-operation of the manostat.

diameter. After the glass tubes were gently removed with hydrofluoric acid the specimens were cut chemically, without introducing any distorsion, to the desired length, and were annealed at 190°C in vacuo for two hours. The specimen No. 1 was a single crystal of diameter 2.4 mm and length 70 mm and the specimen No. 2 was a polycrystalline one of diameter 1.5 mm and length 86 mm; the dimension of crystallites in the specimen No. 2 was comparable with its diameter. Both specimens showed the same critical temperature 3.73_2°K.

III. Results

1. the confirmation of the intrinsic nature of the paramagnetic effect and the investigation of the question concerning the existence of hysteresis phenomena in the effect, by the statical method

All the researchers who gave hitherto valuable contributions to the paramagnetic effect resorted to the method of measuring the magnetization of the specimen by changing any one of three variables I, H and T i.e. the dynamical method, similar in principle to the second method described in the last section. So long as one employs any one of these methods, one may not be able to answer the question that the paramagnetic effect is a transient, apparent phenomenon accompanying the change in time of any one of I, H or T. Therefore we employed the statical method described in the last section in order to see whether the phenomenon is an intrinsic one independent of the measuring procedure or not. In the course of our investigation Teasdale and Rorschach(12) confirmed also the intrinsic nature of the paramagnetic effect which appeared under a definite condition with respect to I and H. Our results obtained by the statical method are shown in Fig. 3 and 4. In Fig. 3 two variables H and T were fixed throughout, while the third one I was changed in small steps. In Fig. 4 I and T were fixed throughout while H was changed in small steps from the value above H_c to zero and vice versa. The search coil C_1 was dropped at each step. The galvanometer deflection was noted and plotted as on Figs. 3 and 4. The same results were obtained whether the directions of I and H were parallel

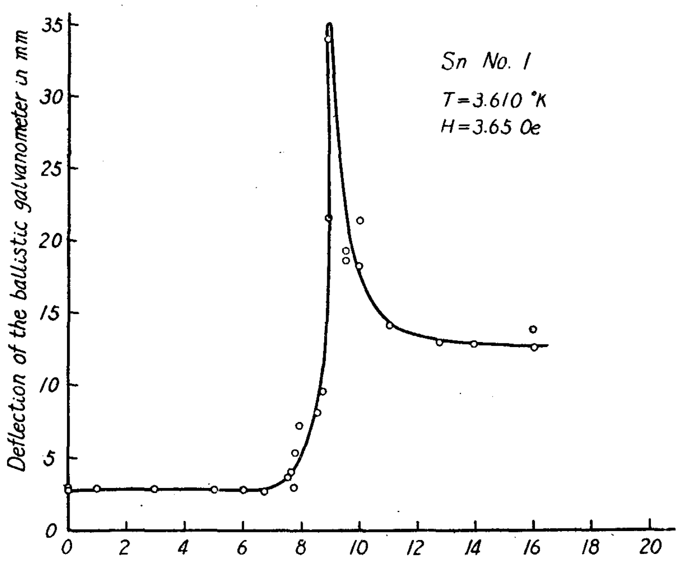

Fig. 3. Galvanometer deflection for No. 1 specimen as a function of current I in the specimen. The coil C_1 was dropped from the lead to the tin section of the specimen.

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(12) T.S. Teasdale and H.E. Rorschach, Phys. Rev., **90** (1953), 709.

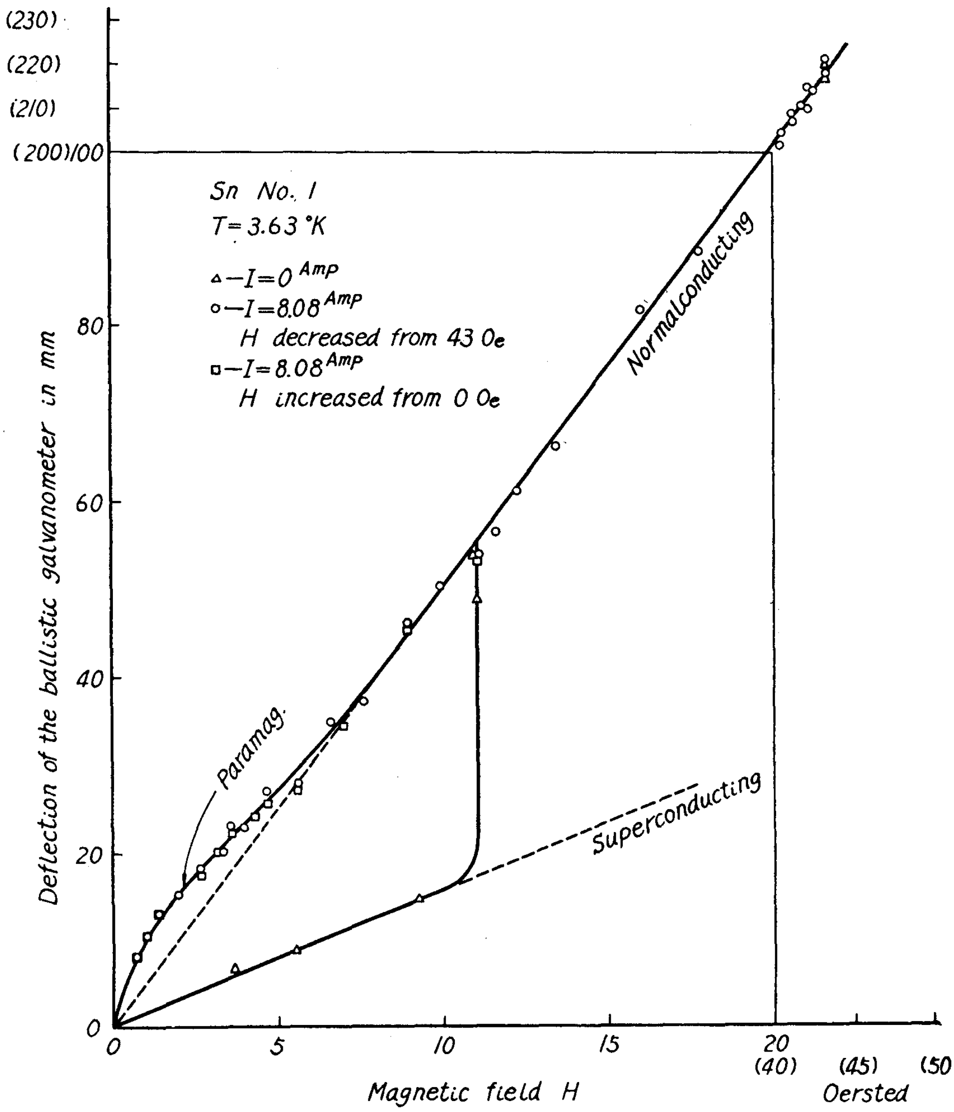

Fig. 4. Galvanometer deflection for No. 1 specimen as a function of external magnetic field H for the case of studying the hysteresis.

or antiparallel to each other. Thus it was ascertained that the phenomenon was not an apparent but an intrinsic one. We can conclude also that there is no hysteresis in the paramagnetic effect and the effect is quite reversible within experimental errors as shown in Fig. 4.

Thompson and Squire(13), who extended the earlier work of Teasdale and Rorschach in detail by the statical method, reported that the paramagnetic effect could be observed, irrespective of the choice of any one of I, H and T as a variable which was changed in small steps, and the effect was completely reversible. Their result is quite in agreement with ours in this respect.

2. Conditions for the appearance of the paramagnetic effect

As described in the introducton, both Steiner(4) who proposed, and Meissner et al.(6) who extended the relation (1) between the current minimum I_0 and the

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(13) J. C. Thompson and C. F. Squire, Phys. Rev., **96** (1954), 287.

external field H, did not take the temperature T into consideration. The formula which define the current minimum should be determined in the three-dimensional (I-H-T) space. For this purpose we employed the dynamical method aforementioned which suffered less fluctuation in measured values than the stasical one. It was at one's disposal which two variables of I, H and T to fix and which one to change in small steps. For convenience' sake, holding T constant throughout, taking I as a parameter and reversing H which was changed in small steps, we measured the ballistic deflection. The typical examples of results obtained with the specimens Nos. 1 and 2 are shown in Figs. 5 and 6(14) respectively. The lines

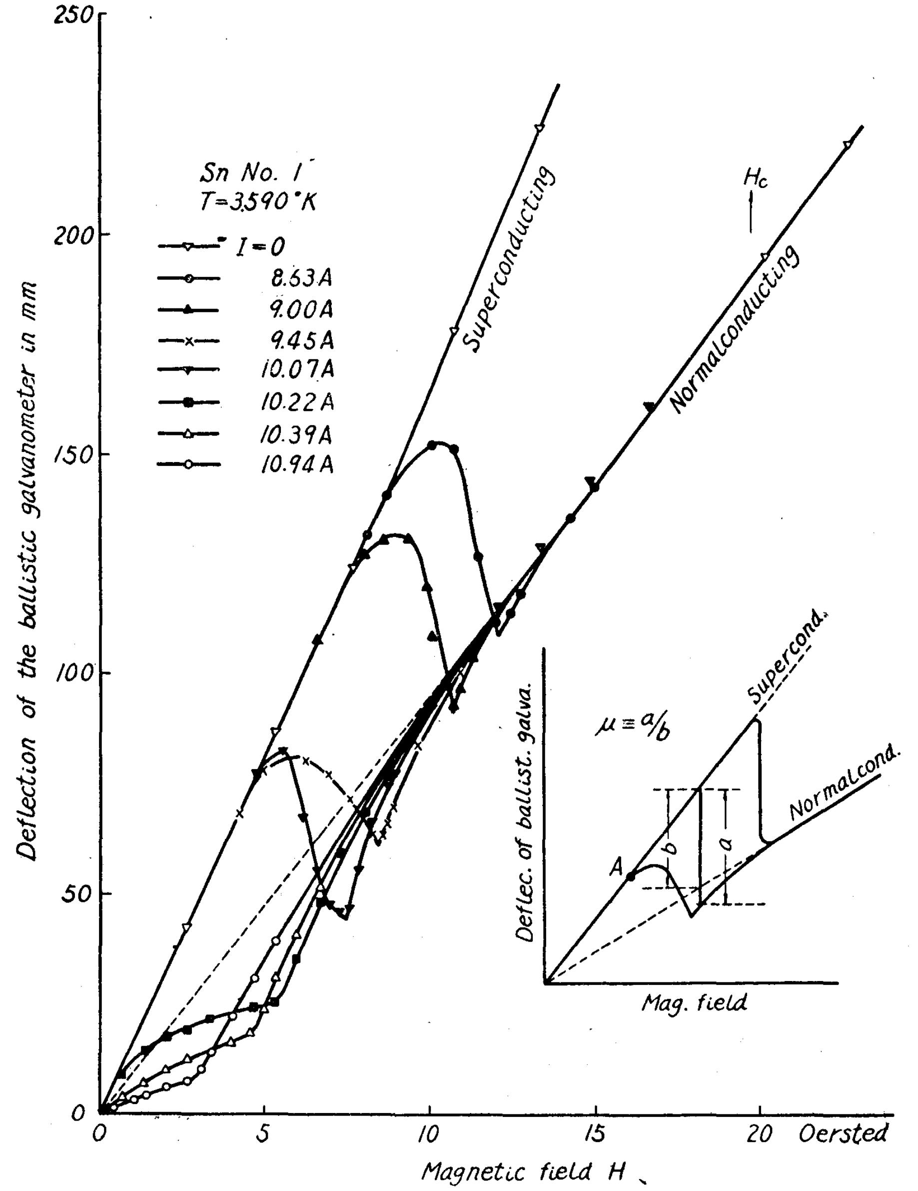

Fig. 5. Galvanometer deflection for No. 1 specimen as a function of external magnetic field H for the specified values of current in the specimen, when H was reversed. a/b shown in the inset gives the apparent permeability μ.

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(14) The installation of the specimen No. 2 in the cryostat was different from that of the specimen No. 1 in that the lead rod Pb used as a thermal valve in the case of the specimen No. 1 was discarded in this case. The relative position of the line "superconducting" tot

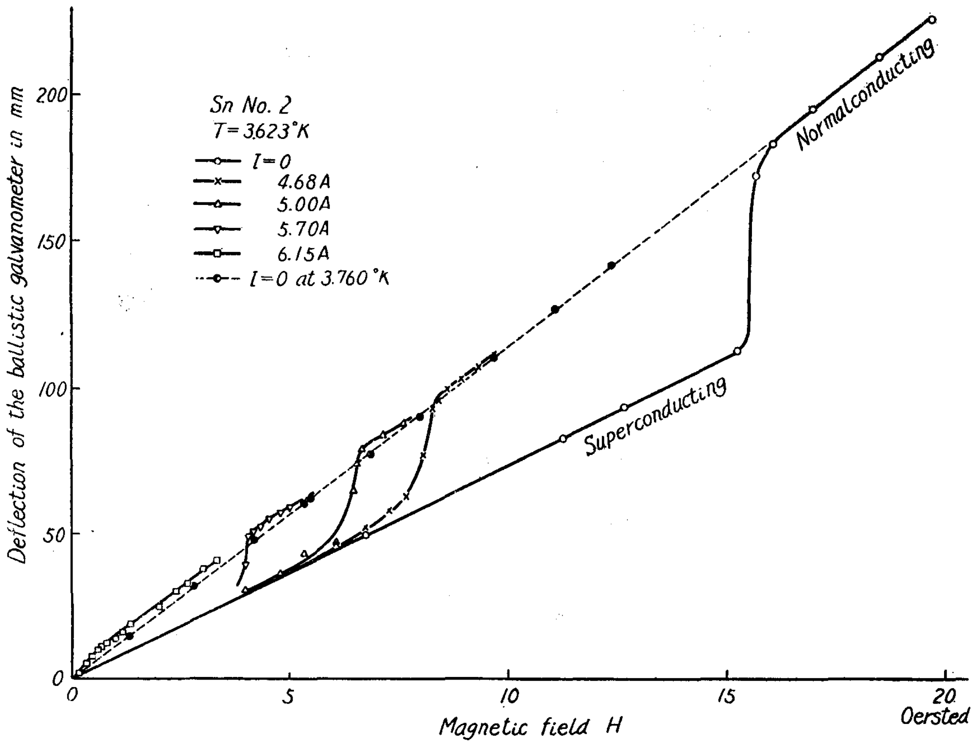

Fig. 6. Galvanometer deflection for No. 2 specimen as a function of external magnetic field H for the specified values of current in the specimen, when H was reversed.

"normalconducting" and „superconducting" in Fig. 5 corresponds to the magnetization curves of the specimen No. 1 in the normal state and in the superconducting state respectively. Owing to the imperfect compensation of the coil C_3 the "normalconducting" line does not coincide with the abscissa but has an inclination to it. Therefore the difference of deflections between the lines "superconducting" and "normalconducting" represents the magnetization of a perfectly diamagnetic body. The measured points which lie under the "normalconducting" line correspond to the paramagnetism. The curve for $I = 10.07$ A crosses the curve for $I = 9.45$ A in the region between the "superconducting" and "normalconducting" lines. Since this may be due to an unexpected temperature change during the measurement perhaps linked to the temporary inactivity of the manostat which might be overlooked, we need not attach great impotance to it. a/b shown in the inset of Fig. 5 gives the apparent permeability μ, which is a function of H provided I and T are constant. (Fig. 7) μ^* which designates the maximum of μ for fixed value of I and T has a meaning similar to that of $\tilde{\mu}$ defined by Meissner et al.(6) μ^* was plotted against I, and the extrapolation to the abscissa for which $\mu^* = 1$ defined the current minimum I_0 at that temperature. Figs. 8 and 9 show these extrapolations at the specified temperatures for the specimens Nos. 1 and 2 respectively. From Figs. 8 and 9 we obtained the $I_0 - T$ relations for two

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†the line "normalconducting" for the specimen No. 2 is contrary to that for the specimen No 1, because in this case C_2 and C_3, used as a search coil and as a compensating coil respectively in the former case, was used as a compensating coil fixed around the copper lead and as a search coil fixed around the center of the specimen No. 2 respectively.

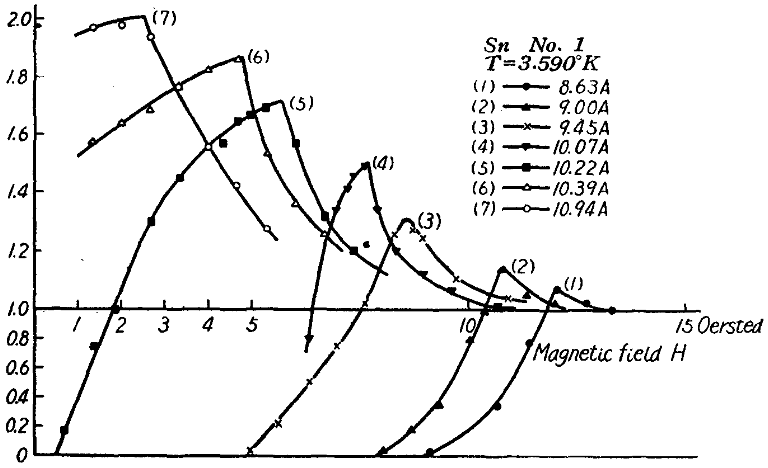

Fig. 7. μ for No. 1 specimen as a function of external magnetic field H for the specified values of current in the specimen at 3.590°K.

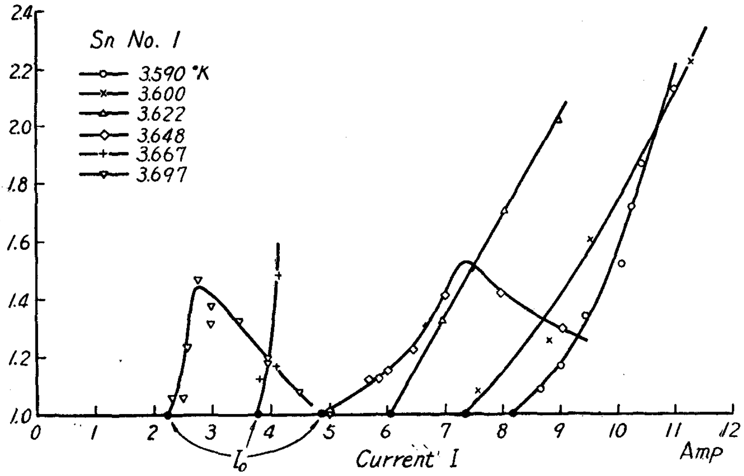

Fig. 8. μ* for No. 1 specimen as a function of current in the specimen at the specified temperatures.

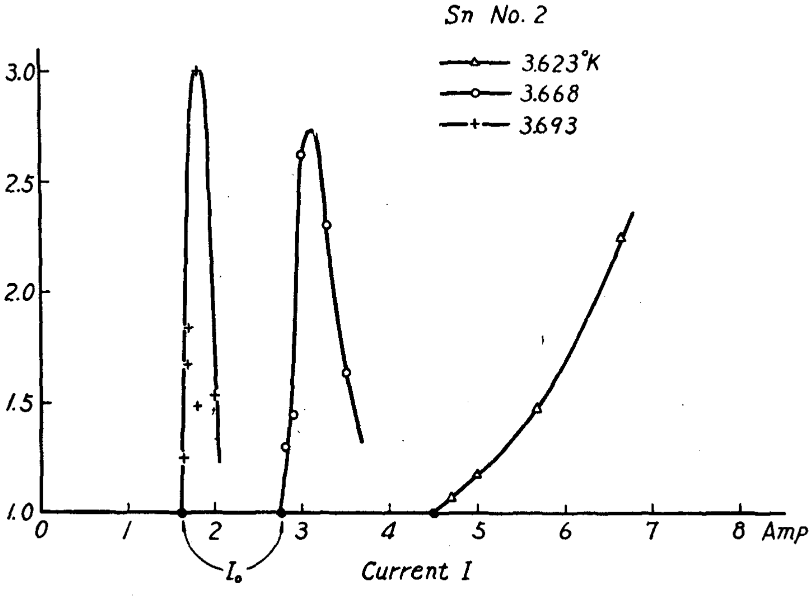

Fig. 9. μ* for No. 2 specimen as a function of current in the specimen at the specified temperatures.

specimens. In a similar way we plot μ^* aginst H^*, the magnetic field which corresponds to μ^* at fixed values of I and T, and obtain H_0, the magnetic field over which we cannot observe the paramagnetism at a given temperature by the extrapolation of μ^* to the abscissa. H_0 changes with the change in T and there is a one-to-one correspondence between H_0 and T (Figs. 10 and 11). Thus we obtained the H_0-T relations. Further we obtained the I_0-H_0 relations for two specimens from Figs. 8, 9, 10 and 11. The I_0-T, the H_0-T and the I_0-H_0 relations thus obtained for two specimens are shown as straight lines at least in the

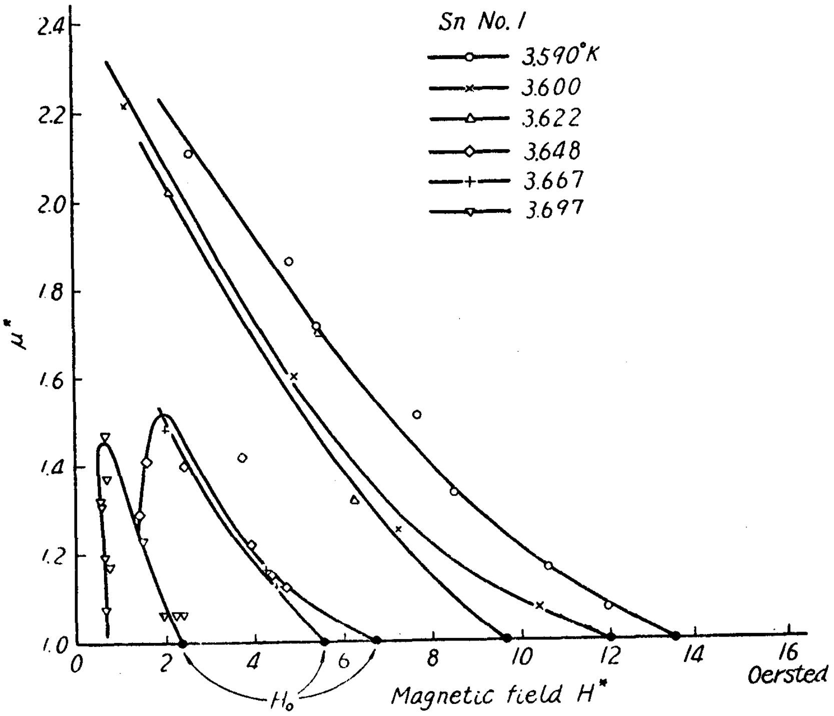

Fig. 10. μ^* for No. 1 specimen as a function of external magnetic field H^* at the specified temperatures.

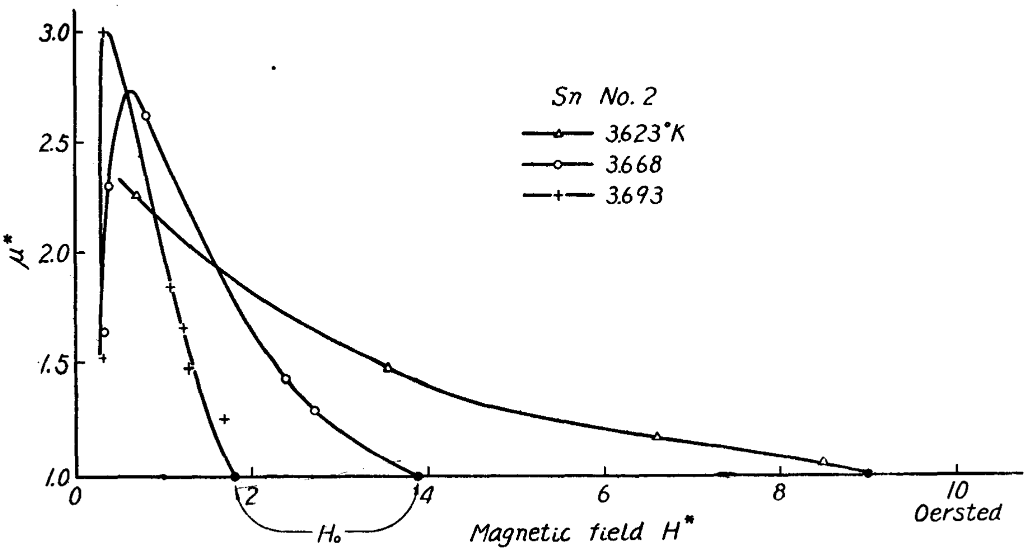

Fig. 11. μ^* for No. 2 specimen as a function of external magnetic field H^* at the specified temperatures.

measured region in Figs. 12, 13 and 14 respectively. Then it was ascertained that the formula for the minimum current requirement was represented graphically by a straight line (the critical line) in the $(I\text{-}H\text{-}T)$ space, the orthogonal projections on the $(I-T)$, the $(H-T)$ and the $(I-H)$ plane of which are the straight lines in Figs. 12, 13 and 14 respectively.

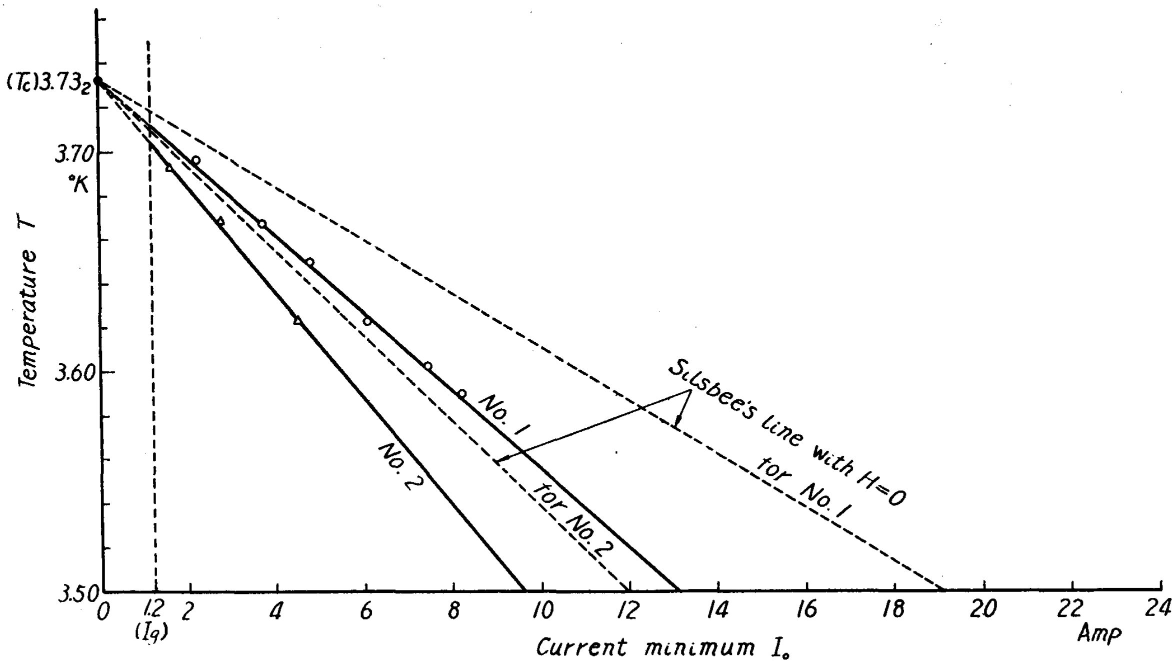

Fig. 12. I_0–T relations for the specimens Nos. 1 and 2.

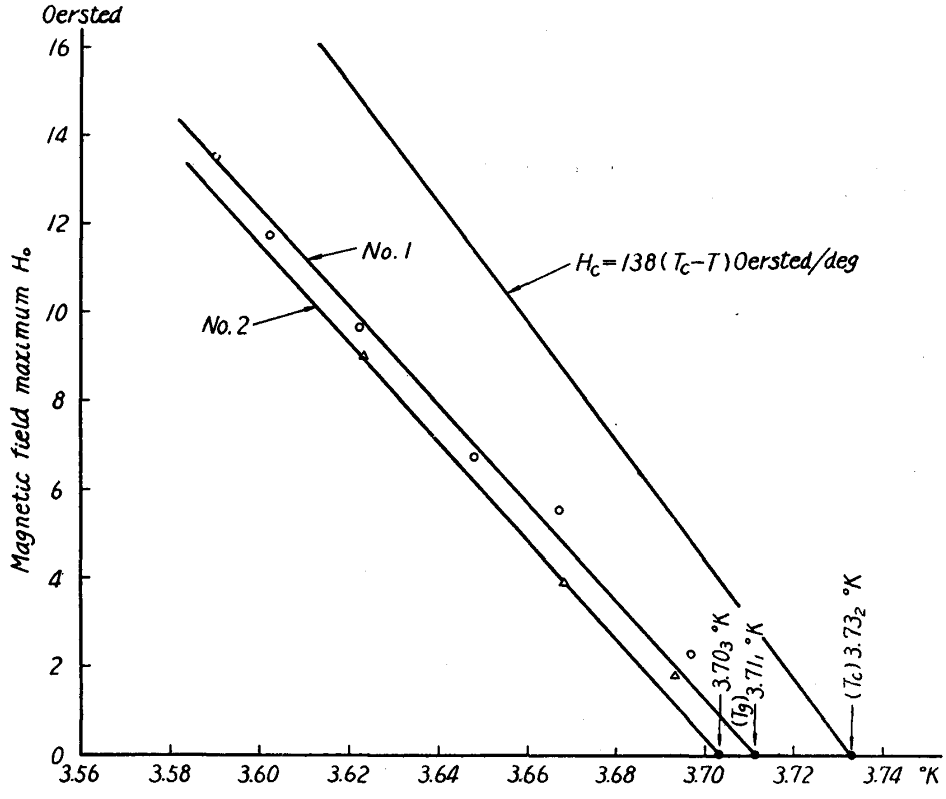

Fig. 13. H_0–T relations for the two specimens.

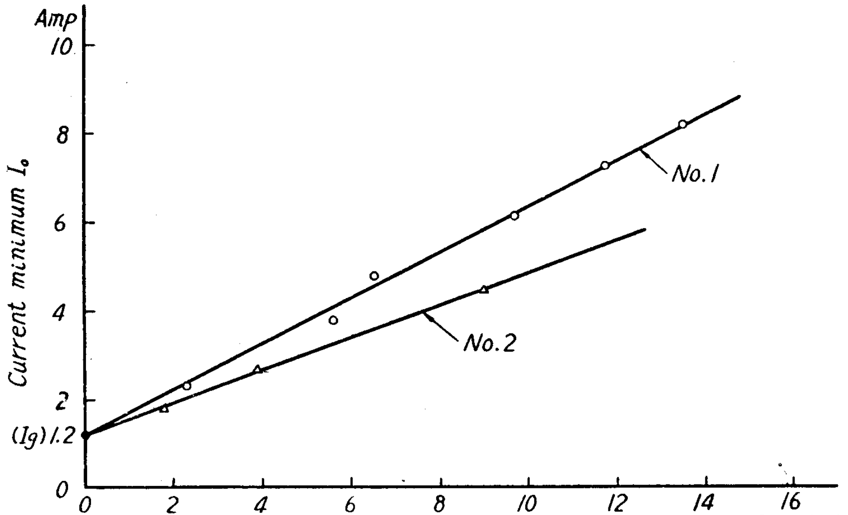

Fig. 14. I_0–H_0 relations for the two specimens.

Though the $(I-T)$ projections of the critical lines have inclinations, proportional to the specimen diameter d, to the T-axis, both the $(I-T)$ projections point to the transition temperature T_c. The $(H-T)$ projections are parallel to each other and T_c-T_g is inversely proportional to d, where T_g is the intersection of the projection with the T-axis. The $(I-H)$ projections have inclinations, proportional to d, to the H-axis but intersect with the I-axis at a constant value of current I_g, irrespective of the specimen diameter. Thus the critical lines terminate at the points (I_g, T_g) on the $(I-T)$ plane.

We obtained as the formulae for the $(I-T)$, the $(H-T)$ and the $(I-H)$ projection which satisfy those relations described above, following three equations (2), (3) and (4) respsctively.

$$I_0 = \xi \gamma d(T_c - T) , \qquad (2)$$

$$H_0 = \xi(T_c - T) - I_g/\gamma d , \qquad (3)$$

$$I_0 = I_g + \gamma d H_0 . \qquad (4)$$

In these equations I_g, γ, T_c and ξ are the characteristic constants of the superconductor. Averaging those values, 3.73₂°K, 1.2 amp, 0.22 and 111 oersted/deg for T_c, I_g, γ and ξ respectively of the specimen No. 1 and 3.73₂°K, 1.2 amp, 0.24 and 112 oersted/deg for those of the specimen No. 2, we determined that $T_c = 3.73_2$°K, $I_g = 1.2$ amp, $\gamma = 0.23$ and $\xi = 1.1 \times 10^2$ oersted/deg for tin. Of three equations (2), (3) and (4) only arbitrary two equations are independent and the formula for

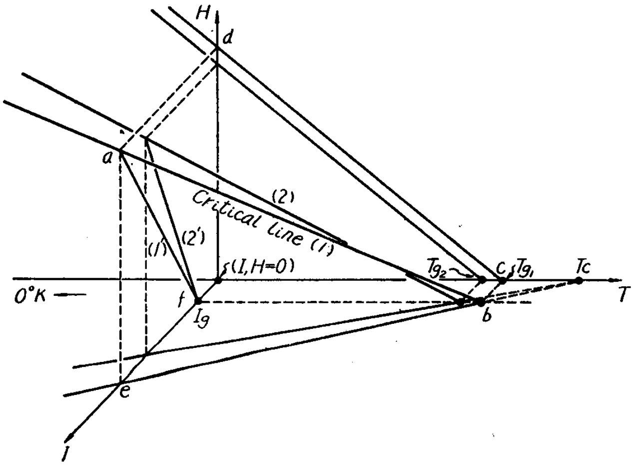

Fig. 15. Critical lines (lines of current minimum) for the specimens Nos. 1 and 2 in the (I–H–T) space. The (I–H) projections (1′) and (2′) of the critical lines are represented well by *Eq.* (1) in the text.

the critical line is represented by the simultaneous equations of the two. Fig. 15 shows schematically the critical lines for two specimens in the $(I-H-T)$ space. The paramagnetism can be observed only in the region of larger I and smaller H than those given by the critical line; for example, in the region under the plane abcd (smaller H) and outside the plane abe (larger I) for the specimen No. 1. The coincidence of *Eqs.* (1) and (4) tells us that the formula obtained by Meissner et al. is not the one for the critical line itself, but the one for its $(I-H)$ projection. Though we obtained 1.2 amp for I_0 in accordance with Steiner(4), and Meissner et al.(6), the value 0.23 for γ obtained by us is not in agreement with the value 0.17 obtained by Meissner et al. It is not clear about the origin of this discrepancy(15).

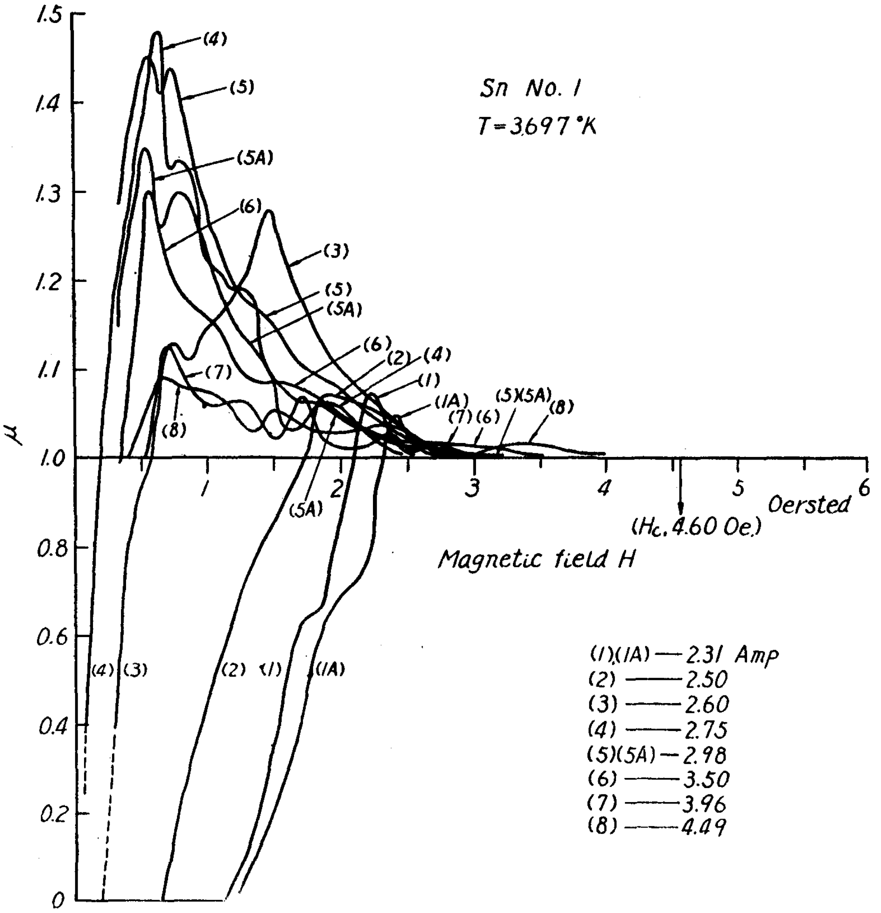

Fig. 16. μ for No. 1 specimen as a function of external magnetic field H for the specified values of current in the specimen at 3.697°K.

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(15) For instance, the following explanation may be plausible in some degree. In an ideal case the magnetization curve for the case $I=0$ should fall abruptly from the "superconducting" line to the "normalconducting" one at H_c at a given temperature. Actually, however, the tail of the magnetization curve does not show an acute angle but becomes somewhat round off. The existence of this small tail for the case $I=0$ may give rise to the overestimation of I_0 and the underestimation of H_0 for which $\mu^*=1$, when a current flows along the specimen. The correct value of γ may be smaller than 0.23. On the other hand, although the procedure of extrapolation after Meissner et al.(6) may be in error in the estimation of α of higher value for a fixed current, the extrapolation to the case for which $\mu=1$ may involve little error in the estimation of I_0 for a fixed value of H.

It should be noted that in some measurements a somewhat systematic structure in the apparent permeability curve can be observed as shown in Fig. 16. The μ curve against H is not such a smooth one as that in Fig. 7 but shows a somewhat periodic deviation from a smooth curve. Each of peaks in the curve seems to be followed systematically from one curve to the next with the change in the parameter I. We observed the systematic structure only when measurements were performed with larger current, at relatively higher temperatures and in such a way that the magnetic field to be reversed was changed in smaller steps than usual. The reproducibility of the fine structure in the μ curve can be seen in comparing the curves (5) and (5A) which were obtained with the same current at the same temperature at different runs. It was very difficult to realize the perfect reproducibility of the structure, owing to the fact that measurements were very sensitive to the change in the temperature and in the current. It may be seen, however, that such a structure is not a nonsensical, random one. The existence of such a structure in the μ curve as shown in Fig. 16, in conjunction with the recent investigation made by Meissner(16), may be one of clues for the study of the mechanism of the paramagnetic effect. We adopted the largest μ as μ^* in such cases.

IV. Paramagnetic region

In order to study the magnitude of the paramagnetic permeability at every point in the paramagnetic region determined in the last section, we plotted for the specimen No. 1 contour lines for which $\mu = \text{const}$ on the $(I-H)$ planes at the temperatures studied. As can be seen in Figs. 7 and 16, which were obtained at 3.590°K and at 3.697°K respectively, the curve which was drawn through the point of μ^* defined in the last section becomes the ridge on the plane concerned. Further at least on the $(I-H)$ planes at higher temperatures near

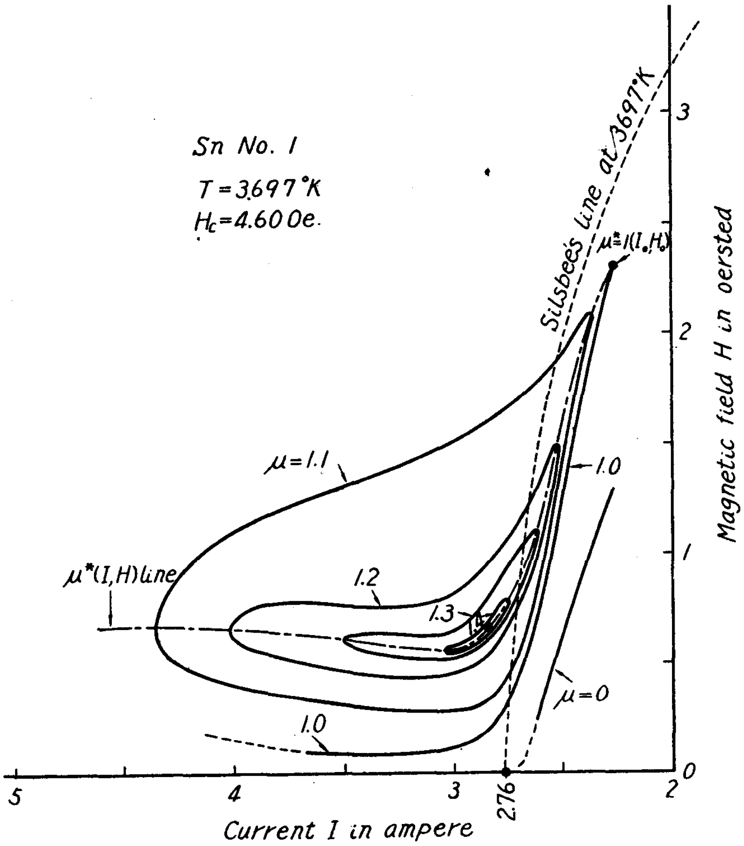

Fig. 17. Contours of μ for No. 1 specimen at 3.697°K.

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(16) W. Meissner, Natwiss., **41** (1954), 437.

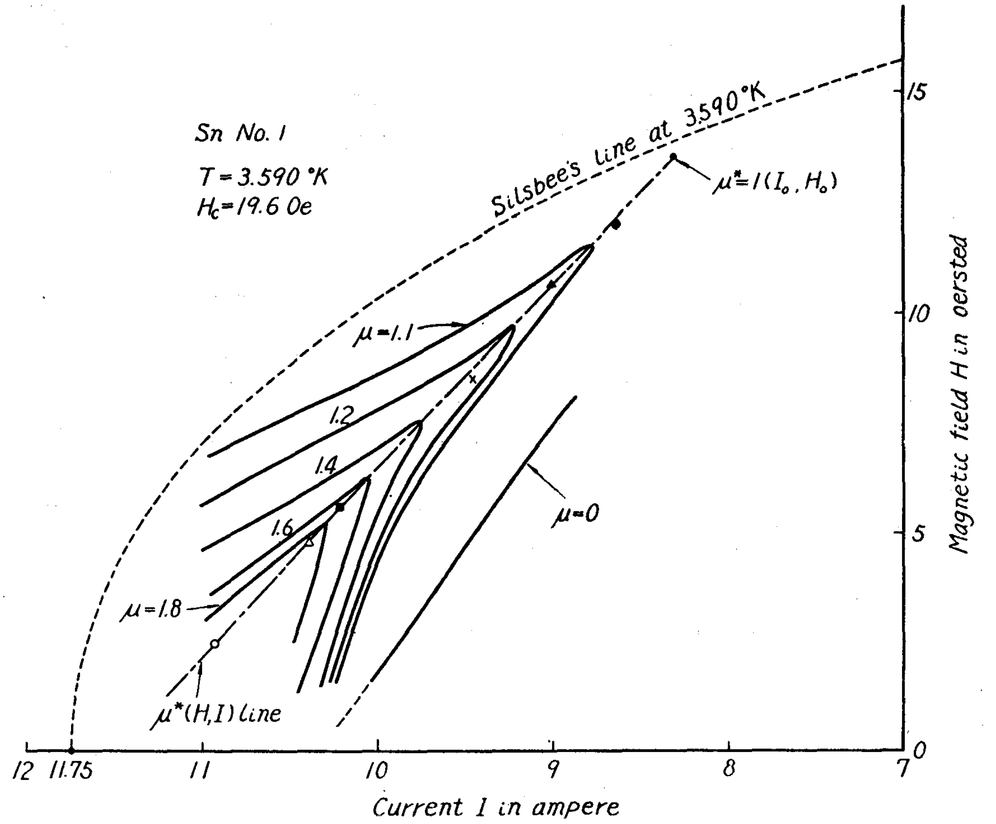

Fig. 18. Contours of μ for No. 1 specimen at 3.590°K.

T_g we obtained closed contour lines which converged with increasing values of μ to a point of finite maximum at finite values of I and H. This can be seen in Fig. 17 obtained at 3.697°K. We could not, however, ascertain whether contour lines closed or not in Fig. 18 obtained at 3.590°K. Fig. 17 and 18 were drawn in such a way as follows; for instance, after finding in Fig. 16 the magnitude such as 2 oersted, 1.5 oersted etc. of the magnetic field H which correspond to $\mu = 1.1$ according to the values of the parameter I such as 2.60 amp, 2.75 amp etc. respectively, we joined with a smooth curve these points (2.60 amp, 2 oersted), (2.75 amp, 1.5 oersted) etc. on the $(I-H)$ plane at that temperature. The curves "$\mu=0$" in Figs. 17 and 18 are what were obtained by connecting the points such as A shown in the inset of Fig. 5 where the magnetization curve branches from the "superconducting" line. The curve designated as Silsbee's lines(17) in the same figures are that which correspond to the formula $\{(4I/cd)^2 + H^2\}^{\frac{1}{2}} = H_c$. It is noteworthy that there exist contours of finite values of μ and even those for which $\mu > 1$ inside Silsbee's lines. Therefore it means perhaps that it is no longer significant to consider that H and H_I are mutually independent and perpendicular to each other in order to get the effective field to destroy the superconductivity and that an unknown mechanism of the paramagnetic effect pushes down Silsbee's line to the

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(17) Assuming the validity of Silsbee's hypothesis, we have the transition surface given by $H_I^2 + H^2 = (4I/cd)^2 + H^2 = H_c^2(T)$ in a three-dimensional $(I-H-T)$ space. Silsbee's line is represented by the above formula at the temperature concerned. According to this assumption the permeability should be zero inside this line.

curve "$\mu = 0$". The conspicuous region of paramagnetism is shown stereographically in Fig. 19.

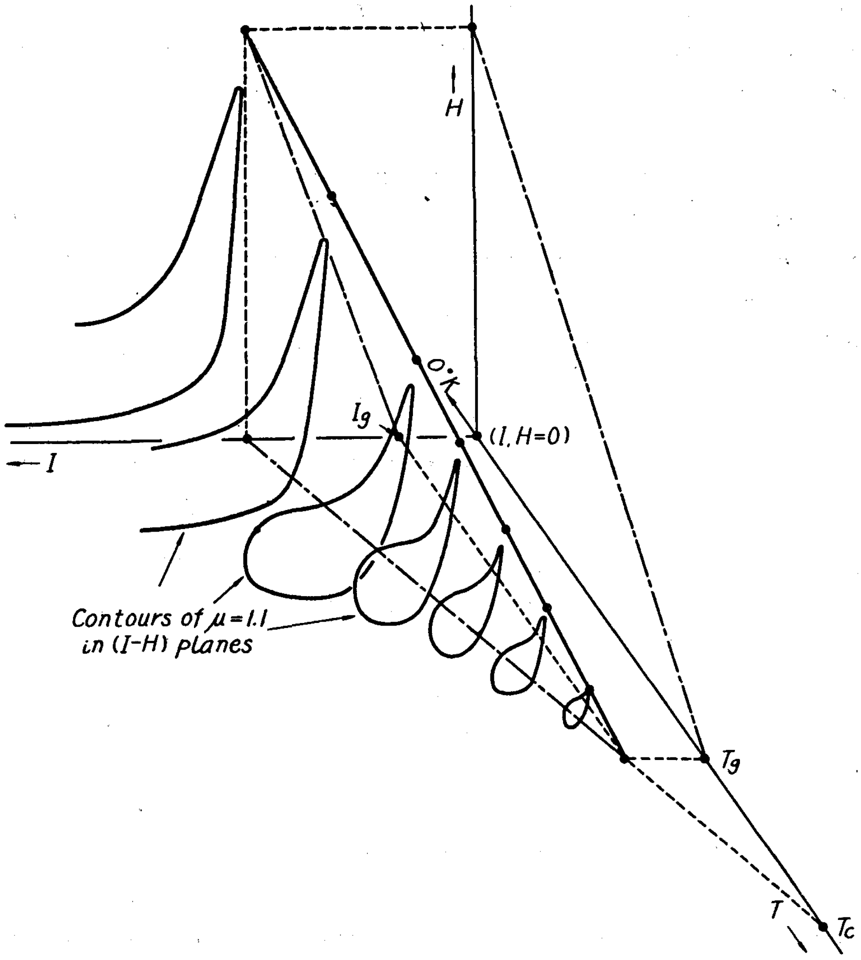

Fig. 19. Conspicuous region of paramagnetism ($\mu \geqq 1.1$) in the three-dimensional space.

Meissner et al.⁽⁶⁾ reported that in some case they had observed the paramagnetism even in the field larger than the critical field H_c. We could not, however, observe such a fact as they did. It is clear from *Eq.* (3) or in Fig. 13 that the $(H-T)$ projections of the critical lines lie under the threshold field curve⁽¹⁸⁾ for tin. A simple extrapolation of the $(H-T)$ projections, which were obtained in the present investigation at relatively high temperatures near T_c, to lower temperatures allows the $(H-T)$ projections to exceed the threshold curve. As it seems plausible to suppose simply that a superconductor may be in the normal state in a magnetic field larger than H_c, even if a current flows along the superconductor, it is not convincible to observe the paramagnetic effect in such a field as larger than H_c. It may be, however, very hasty to deny at once such a result. Therefore it seems worthwhile to examine the result reported by Meissner et al. from this point of view.

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(18) The threshold field curve for tin can be represented well by the straight line $H_c = \xi'(T_c - T)$ at relatively higher temperatures near T_c. As ξ' is 138 oersted/deg, ξ' is larger than ξ.

Moreover, it may be more pertinent to take *Eqs.* (2), (3) and (4) to be an approximation at relatively higher temperature near T_c of formulae including terms of higher order in T, the curve for which would not intersect the threshold field curve at lower temperatures, because our experiments was perfomed only at the temperatures near T_c(19) where the threshold field curve was represented in a good approximation by a straight line.

Setting aside these questions and assuming the validity of *Eqs.* (2), (3) and (4) down to the absolute zero of temperature, it is deduced from *Eqs.* (2) and (3) that we cannot observe the paramagnetism in case that I_0 at 0°K which is equal to $\xi\gamma dT_c$ is less than I_g. It means that there is a lower limit of the specimen diameter from the condition that d is larger than or equal to $I_g/(\xi\gamma T_c)$. For tin the lower limit d_0 is 1.3×10^{-2} mm.

V. Discussions

As touched briefly in the introduction, we have not yet a satisfactory theory on the paramagnetic effect. According to Steiner(4), the paramagnetic effect could be explained qualitatively with Stark's working hypothesis(20) on superconductivity. The working hypothesis seems, however, to be out of date at present. Steiner concluded, moreover, from the result that he could not observe the paramagnetic effect with mercury, that the effect could be observed only with such superconductors as had p electrons in the outermost shell of atoms of which the metal consisted. Since Meissner et al.(6) discovered afterwards the paramagnetism also in mercury, this view became an unauthentic one.

Meissner et al.(7) proposed an idea that in the intermediate state the specimen-current following a helical path so as to connect the oblong superconducting grains oriented along the resultant field (current field plus external field) produces a longitudinal flux greater than that due to the external field. According to this idea such a helical current produces always the paramagnetism in a cylindrical specimen in an external longitudinal field, whether the directions of the current and the external field are parallel to each other or antiparallel. Meissner et al. remarked that it could not, however, explain the paramagnetism observed with a hollow cylindrical superconductor in an external circular field perpendicular to the specimen axis.

Recently H. Meissner formulated a theory(21), basing upon the idea due to Meissner et al. Although this theory is not able to explain what happens except at such a peak value μ^* (k_m in his paper) as shown in Figs. 7 and 16, the agreement of the theory with the experiment is fairly good. According to H. Meissner,

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(19) The increase of necessary current for the paramagnetism with the decrease of temperature makes it hard to perform an experiment at lower temperatures, b cause a development of a large amount of Joule's heat attendant on the increased current makes it very difficult to keep the temperature constant within the specimen.

(20) J. Stark, Phys. Z., **36** (1935), 515, **38** (1937), 269.

(21) H. Meissner, Phys. Rev., **97** (1955), 1627.

it seems that one should expect μ^* to be on Silsbee's lines shown in Figs. 17 and 18. His opinion seems to agree with that of Thompson and Squire(13) in this respect. If it were true, we consider that Silsbee's line should coincide not only with the line μ^* but also with the line "$\mu = 0$" in the same figures. So far as the present investigation concerns, it seems unlikely that Silsbee's line coincides with the lines μ^* and "$\mu = 0$". Even if one takes into account the heating due to a current which may cause the temperature in the specimen to be slighly higher than the temperature of the liquid helium bath, one may be not to expect simply the exact coincidence of Silsbee's line and, the lines μ^* and "$\mu = 0$". Another possibility thought of(22) is that the critical line may be the intersection of the transition surface afore-mentioned and the plane given by *Eq.* (4). If this suggestion were true, the $(I-T)$ projection of the critical line shoud not point to the critical temperature T_c. This is not in agreement with our results. Furthermore, the characteristic constant I_g cannot be explained with H. Meissner's theory. We believe at present we can say nothing about μ^* lines in Figs. 17 and 18 and I_g without a more exact knowledge of the mechanism of the flux increase.

Assuming that the total current followed a helical path on the surface of the tin specimen, Thompson and Squire(13) showed that the total turn number a certain current made in passing through the specimen was nearly equal to the number of revolutions made by an electron with a cyclotron angular velocity given by the longitudinal field, which travelled with the velocity at the top of the Fermi distribution from one end of the specimen to the other without suffering collisions. Such a model is not, however, plausible in principle, because a cyclotron angular velocity proposed by them is to give rise not to a paramagnetic orbit, but to a diamagnetic one. Even if one takes the reflections of electrons at the specimen surface into consideration, it cannot be concluded from a speculation based upon the classical concept of orbital motions of eletrons, whether one expects paramagenetic orbits for electrons.

Thus the existing ideas and theory cannot explain fully our experimental results. We are continuing the investigation in a wider field of vision in order to have a more extensive knowledge of the paramagnetic effect. Experiments on tin specimens having cross-sections different from circular and on other superconductors such as mercury and indium are in progress.

We would like to express our thanks to Professor T. Fukuroi who gave us facilities for experiments and helpful suggestions.

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(22) J. C. Thompson gave us the same suggestion in a private communication.