Type I to Type II Transition in Superconducting In-3.9 at.%Pb Alloy

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Type I to Type II Transition in Superconducting In-3.9 at.%Pb Alloy*

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Synopsis

The magnetization and the thermal conductivity of In-3.9 at.% Pb alloy are measured between 1°K and 4.2°K. It is found that this alloy sample does show type I to type II transition of the superconductivity at $T_{cr}=2.2°K$. Below T_{cr}, the magnetization decreases with finite slope near the upper critical field, H_{c2}, and the thermal conductivity shows deep minima both in the magnetic field dependence and in the temperature dependence. Above T_{cr}, on the other hand, the sample does not show any such effects mentioned above.

The data concerning the residual resistivity $\rho_n=2.46\mu\Omega$cm, the transition temperature $T_c=3.64°K$, the mean free path $l=5.69\times10^{-6}$cm, the G-L parameters $\kappa_1(0)=0.77$ (at 0°K), $\kappa_1(1)=0.68$ (at T_c) and the ratio $\kappa_1(0)/\kappa_1(1)=1.13$ are obtained. These values are all in good agreement with the results of previous investigations on In-Pb alloys.

1. Introduction

There have been many experiments on type II superconductivity which is described by Ginzburg-Landau-Abrikosov theory.(1),(2) After this theory, it has turned out obvious as a result of theoretical works by Gor'kov,(3) Maki,(4) Helfand and Welthamer,(5) Eilenberger,(6) et al. that Ginzburg-Landau (G-L) parameter, $\kappa_1(t)$, increases with decreasing temperature. According to Maki's theory,(4)

$$\frac{\kappa_1(0)}{\kappa_1(1)}=1.25\,, \qquad \text{for pure limit } (l\gg\xi_0) \tag{1}$$

and

$$\frac{\kappa_1(0)}{\kappa_1(1)}=1.20\,, \qquad \text{for dirty limit, } (l\ll\xi_0) \tag{2}$$

where t, l and ξ_0 denote the reduced temperature, the mean free path and the coherence length of electrons, respectively. The behavior that $\kappa_1(t)$ increases with decreasing temperature has been verified with many metals and alloys. Thus

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* The **1382nd** report of the Research Institute for Iron, Steel and Other Metals.

(1) V.L. Ginzburg and L.D. Landau, Zh. eksper. teor. Fiz., **20** (1950), 1064.

(2) A.A. Abrikosov, Soviet Physics-JETP, **5** (1957), 1174.

(3) L.P. Gor'kov, Soviet Physics-JETP, **10** (1960), 593.

(4) K. Maki, Physics, **1** (1964), 21 and 127.

(5) E. Helfand and N.R. Welthamer, Phys. Rev. Letters, **13** (1964), 686.

(6) G. Eilenberger, Phys. Rev., **153** (1966), 584.

some superconductors with κ-values of slightly smaller than $1/\sqrt{2}$ at the transition temperature, T_c, should show the transition from type I to type II as the temperature is lowered below the critical temperature, T_{cr}, where $\kappa_1(T_{cr})=1/\sqrt{2}$. Such a substance has never been found in pure metals. Nb, V and Tc show type II superconductivity throughout the superconducting range inasmuch as their $\kappa_1(1)$-values being larger than $1/\sqrt{2}$, while most of the other metal superconductors show type I behavior in their whole superconducting temperature range as they have $\kappa_1(0)$-values smaller than $1/\sqrt{2}$.

In the case of alloy superconductor, $\kappa_1(1)$ depends on the mean free path and is expressed by Gor'kov-Goodman,(7),(8) as follows:

$$\kappa_1(1)=\kappa_0+a\rho_n\gamma^{1/2}, \tag{3}$$

where κ_0, ρ_n and γ are the intrinsic G-L parameter, the residual resistivity and the electronic specific heat coefficient, respectively. Thus one can prepare an alloy superconductor which shows the transition from type I to type II by adjusting the concentration of impurity.

Physical properties such as the magnetization, the thermal conductivity, the specific heat differ remarkably whether the sample belongs to type I or type II. In the type I superconductor, the magnetization curve discontinuously drops at thermodynamic critical field, $H_c(t)$. In the type II superconductor, on the other hand, the magnetization continuously decreases in the mixed state and vanishes with finite slope at the upper critical field, $H_{c2}(t)$. In the thermal conductivity *vs.* magnetic field curve of type II superconductor, a sharp minimum has been reported(9),(10) in the mixed state region.

Type I to type II transition of superconductivity has been observed by Kinsel et al.(12) for In–1.5 at.% Bi alloy by magnetization measurements. However, no detailed information has been reported.

In a previous work(11) (which we shall refer to I hereafter), the magnetization of In alloys containing 4.4 at.% to 5.6 at.% Pb were studied as a function of magnetic field and it was predicted that In alloy containing 3.5 to 4.1 at.% Pb should show the transition from type I to type II. The purpose of this work is to confirm that an alloy sample, In–3.9 at.% Pb, shows type I to type II transition at the critical temperature, T_{cr}, from studies of magnetization and thermal conductivity.

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(7) L.P. Gor'kov, Soviet Physics-JETP, **9** (1959), 1364.
(8) B.B. Goodman, Phys. Letters, **1** (1962), 215.
(9) L. Dubeck, P. Lindenfeld, E.A. Lynton and H. Rohrer, Phys. Rev. Letters, **10** (1963), 98; Rev. Mod. Phys. **36** (1964), 110.
(10) T. Mamiya, J. Phys. Soc. Japan, **21** (1966), 1032.
(11) K. Noto, Y. Muto and T. Fukuroi, J. Phys. Soc. Japan, **21** (1966), 2122.
(12) T. Kinsel, E.A. Lynton and B. Serin, Rev. Mod. Phys., **36** (1964), 105.

2. Sample and experimental procedures

The polycrystalline alloy sample of In–3.9 at.% Pb was prepared by the same technique as described in I. It is a rod of 3.5 mm in diameter and 5 cm in length. The residual resistivity was measured after magnetic and thermal conductivity measurements and was determined to be 2.46 $\mu\Omega$cm. This value is in good agreement with the results in I within an experimental accuracy, as shown in Fig. 1.

The magnetization was measured at temperatures between 1°K and 4.2°K by the same cryostat and the superconducting solenoid which were shown as the "apparatus B" in I.

The thermal conductivity was measured by the method previously reported by Mamiya.(10) The cryostat was slightly modified and is shown schematically in Fig. 2. The cryostat can with a smaller diameter was used and another superconducting solenoid which gives more homogeneous longitudinal magnetic fields up to 8 KOe was employed. Two Allen-Bradley's 1/10 watt, 10 ohms carbon resistor thermometers were calibrated for every run and the following expression,(13)

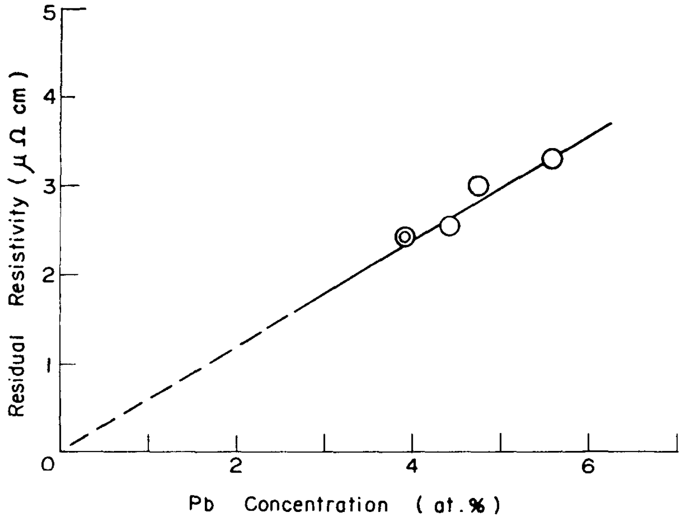

Fig. 1. Concentration dependence of residual resistivity in In-Pb alloy system.
◎: present work, ○: previous work (I).

$$\log R + \frac{A}{\log R} = B + \frac{C}{T}, \tag{4}$$

was used and the constants were determined by means of the method of least squares. All numerical calculations were carried out by using NEAC 2230 Computer, Tohoku University.

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(13) J.R. Clement and E.H. Quinell, Rev. Sci. Instrum., **23** (1952), 213.

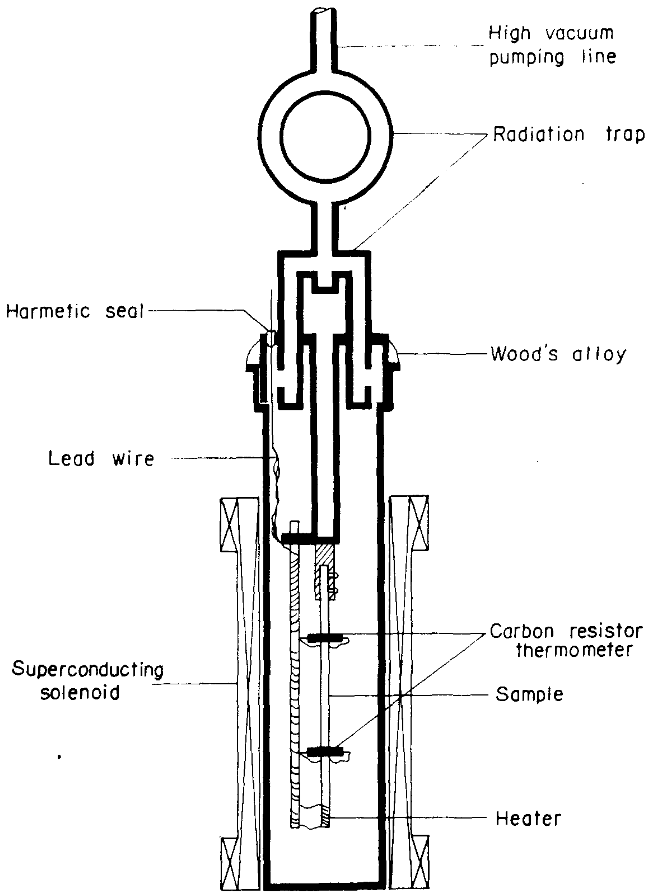

Fig. 2. The cryostat for thermal conductivity measurements shown schematically.

3. Results

A. Magnetization

Figure 3 shows typical magnetization curves in ascending field at 3.01°K and 1.05°K. It can be seen that the magnetization drops with infinite slope at H_c as seen in Fig. 3(a), while it decreases with finite slope near H_{c2} as seen in Fig. 3(b). As described in § 1, these are the typical behavior of type I and type II superconductor, respectively. Only magnetization curve in ascending field was analyzed as explained in I. Thus, lower and upper critical fields, H_{c1} and H_{c2}, were determined as shown in Fig. 3(b), and thermodynamic critical field, H_c, was determined by the following relation,

$$\frac{H_c^2(t)}{8\pi}=\int_0^{H_{c2}}-MdH\,. \tag{5}$$

The transition temperature was determined to be 3.64°K from H_c *vs.* temperature curve. This value is in good agreement with the previous results as shown in Fig. 4.

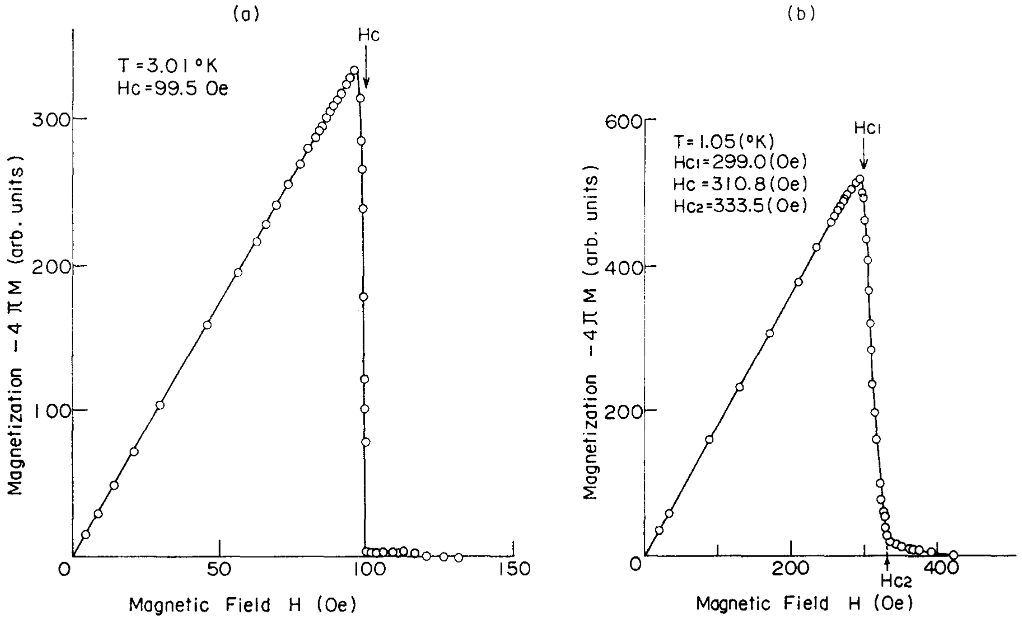

Fig. 3. Magnetization curves. (a): at 3.01°K. (b): 1.05°K.

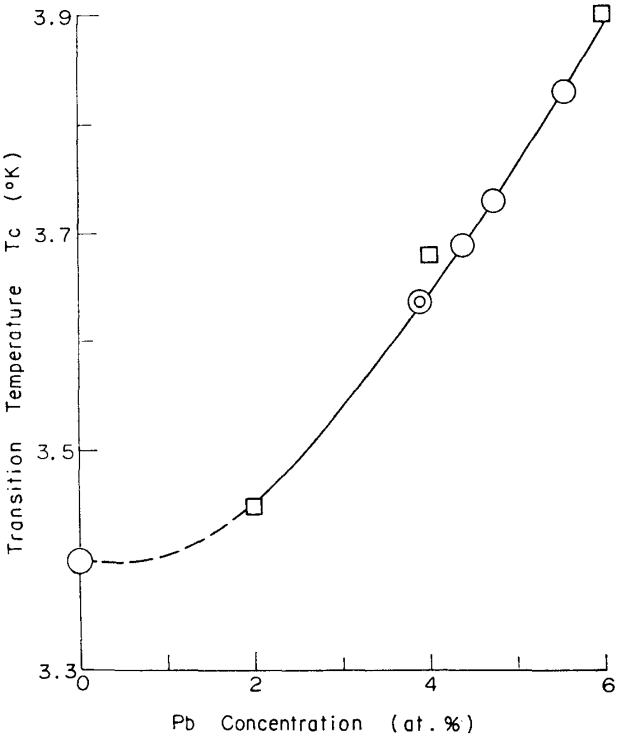

Fig. 4. Concentration dependence of transition temperature in In-Pb alloy system.
◎: present work, ○: previous work (I), □: by Gygax and Kropschot.(14)

Figure 5 shows the temperature dependence of critical fields plotted against the reduced temperature. The critical temperature where both $H_{c1}(t)$ and $H_{c2}(t)$ coincide to $H_c(t)$ is about 2.2°K $[t_{cr}=T_{cr}/T_c=0.605]$.

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(14) S. Gygax and R.H. Kropschot, Phys. Letters, **12** (1964), 7.

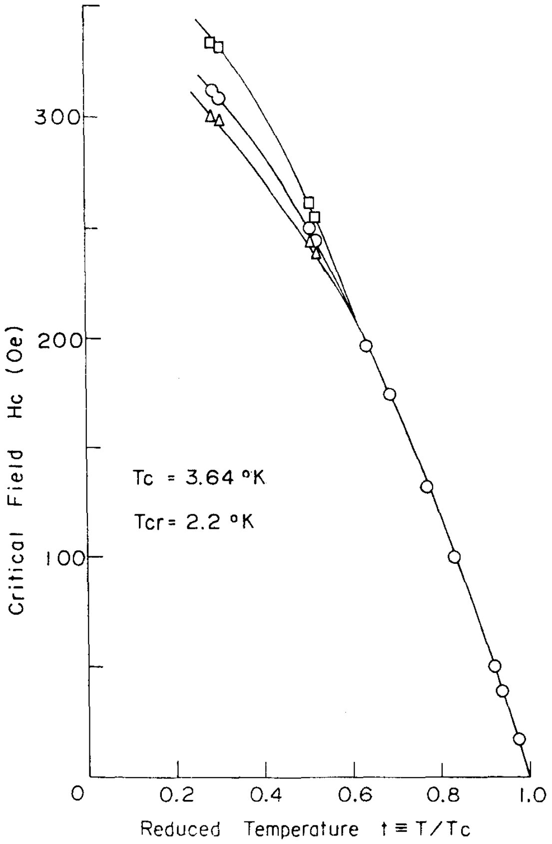

Fig. 5. Temperature dependence of critical fields plotted against $t \equiv T/T_c$. ○: thermodynamic critical field H_c, □: upper critical field H_{c2}, △: lower critical field H_{c1}.

Below T_{cr}, $\kappa_1(t)$ and $\kappa_2(t)$ can be obtained by Maki's definitions[3] as follows,

$$\kappa_1(t) = \frac{H_{c2}(t)}{\sqrt{2}H_c(t)}, \tag{6}$$

$$\left(-4\pi\frac{dM}{dH}\right)_{near\ H_{c2}} = \frac{1}{\beta(2\kappa_2^2(t)-1)}, \qquad \beta = 1.16 \tag{7}$$

Figure 6 shows the temperature dependence of $\kappa_1(t)$ and $\kappa_2(t)$. Here again, one can see T_{cr} to be about 2.2°K where $\kappa_1(t)$ is equal to $1/\sqrt{2}$. The values of $\kappa_1(t)$ and $\kappa_2(t)$ agree with each other within experimental accuracy and increase with decreasing temperature below 2.2°K.

B. Thermal conductivity

Type I to type II transition was confirmed also from the measurements of the thermal conductivity. The thermal conductivity was measured as shown by arrows in Fig. 7. Figure 8 shows the magnetic field dependence of the thermal conductivity at 3.16°K, (① in Fig. 7) and at 1.74°K (② in Fig. 7). It can be seen in

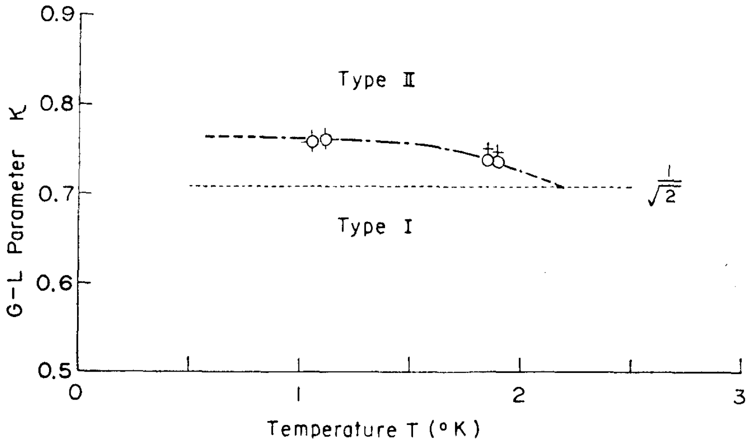

Fig. 6. Temperature dependence of G-L parameters $\kappa_1(t)$ and $\kappa_2(t)$. ○: $\kappa_1(t)$, +: $\kappa_2(t)$.

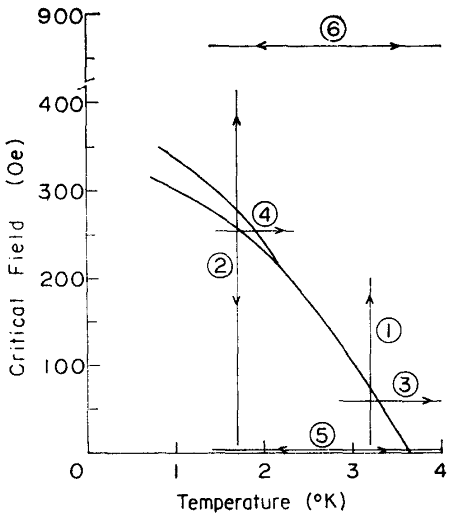

Fig. 7. Temperature dependence of critical fields is shown schematically. Thermal conductivity was measured as shown by arrows ① to ⑥.

Fig. 8(b) that there exists a sharp minimum in the mixed state region between H_{c1} and H_{c2}.(*1) whereas no such anomaly can be seen in Fig. 8(a).

Figure 9 shows the temperature dependence of the thermal conductivity in several magnetic fields. Similar behaviors can also be observed: a sharp minimum appears in the mixed state region between T_1 and T_2, as shown by circles (④ in Fig. 7), where H_{c1} and H_{c2} is equal to 259 Oe, respectively, no anomaly can be seen as shown by triangles at T_H where H_c is 57.6 Oe. (⑤ in Fig. 7)

(*1) The magnetic field dependence of the thermal conductivity just below H_{c2} at 1.74°K (Fig. 7(b)) was found to be in good agreement with the Caroli-Cyrot(15) theory by us.(16)

(15) C. Caroli and M. Cyrot, Phys. kondens. Materie, **4** (1965), 285.

(16) Y. Muto, K. Noto, T. Mamiya and T. Fukuroi, J. Phys. Soc. Japan, **24** (1968), 992.

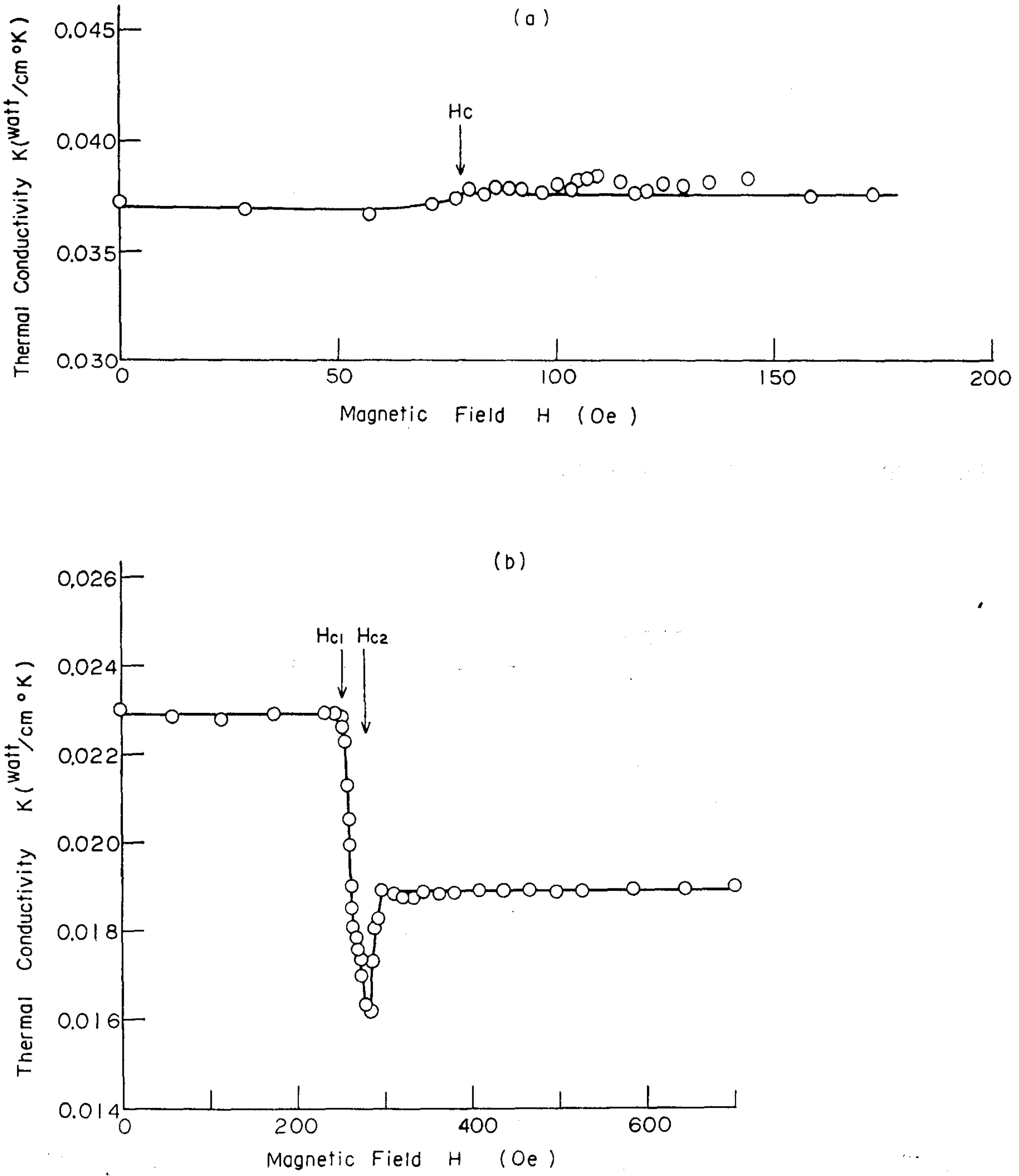

Fig. 8. Mangetic field dependence of the thermal conductivity. (a): at 3.16°K, (b): 1.74°K.

In Fig. 9, the thermal conductivity in normal state was obtained in the magnetic field, $H=864$ Oe, as shown by dashed curve (⑥ in Fig. 7), and that in superconducting state was obtained in $H=0$ as shown by solid curve (⑤ in Fig. 7).(*2)

(*2) In Fig. 8, the thermal conductivity in normal state (shown by dashed curve) is slightly concave upwards with respect to the temperature T because it contains not only electronic part linear in T (limited by impurity scattering) but also phonon part proportional to T^2 (dominated by electron scattering). The reason why the thermal conductivity in superconducting state (shown by solid curve) becomes larger than in normal state may be that the increase in phonon conduction which is due to gradual reduction of normal electron dominates the decrease in electronic conduction more and more with decreasing temperature in superconducting state.

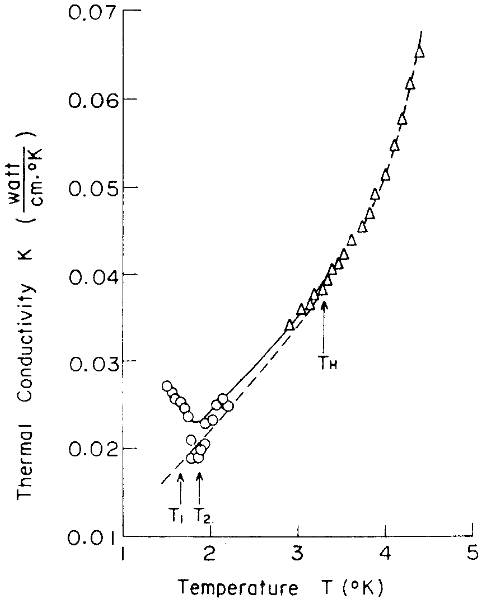

Fig. 9. Temperature dependence of the thermal conductivity in the fixed magnetic field; solid curve: $H=0$ (superconducting), dashed curve: $H=864$ Oe (normal), ○: $H=259$ Oe, △: $H=57.6$ Oe.

4. Discussion

As already mentioned in §3, in the magnetization curve, there is a mixed state region at lower temperature as shown in Fig. 3(b), while there is no such a region at higher temperature as seen in Fig. 3(a). Moreover, a sharp minimum both in the magnetic field dependence and in the temperature dependence of the thermal conductivity were recognized as seen in Fig. 7(b) and in Fig. 8 (shown by circles), while no such anomaly was found in Fig. 7(a) and Fig. 8 (shown by triangles). Thus the transition of the superconductivity from type I to type II was verified by measurements on the magnetization and the thermal conductivity.

The mean free path l and the ratio ξ_0/l of the sample is evaluated to be 5.69×10^{-6} cm and 7.7, respectively, as the measure of dirtiness, following the procedure shown in the previous work (I).

The electronic specific heat coefficient γ of the sample is estimated to be 1.49×10^{3} erg cm^{-3} °K^{-2} with the following BCS relation,⁽¹⁷⁾

$$\frac{\gamma T_c^2}{H_c^2(0)} = 0.17. \tag{8}$$

We can evaluate the value of $\rho_n\gamma^{1/2}$ for the sample to be $0.95\times10^{-4}\,\Omega\text{cm}^{-1/2}\,\text{erg}^{1/2}\,{}^\circ\text{K}^{-1}$ in terms of γ and ρ_n. Thus, we can find the value of $\kappa_1(1)$ to be 0.68 which is smaller than $1/\sqrt{2}$ as shown in Fig. 10. The value of $\kappa_1(0)$ can be obtained to be about 0.77 which is larger than $1/\sqrt{2}$ by the extrapolation to 0°K in Fig. 6. These

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(17) J. Bardeen, L.N. Cooper and J.R. Schrieffer, Phys. Rev., **108** (1957), 1175.

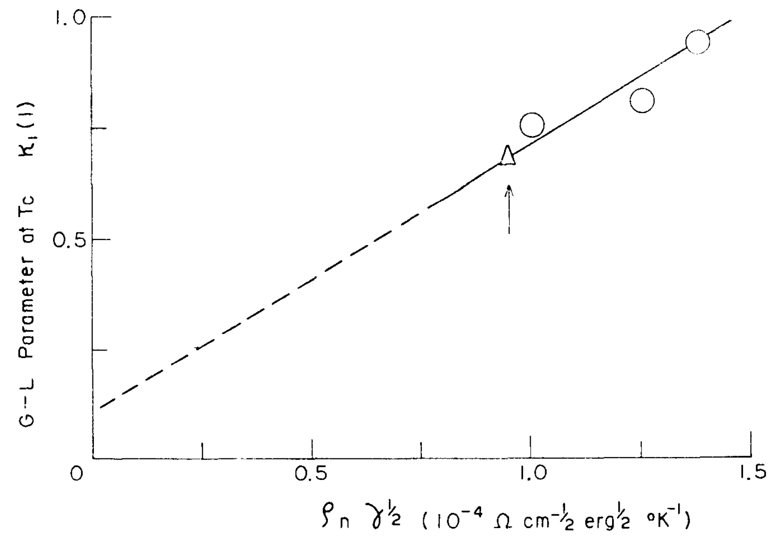

Fig. 10. The variation of $\kappa_1(1)$ with $\rho_n\gamma^{1/2}$. △: present work, ○: previous work (I).

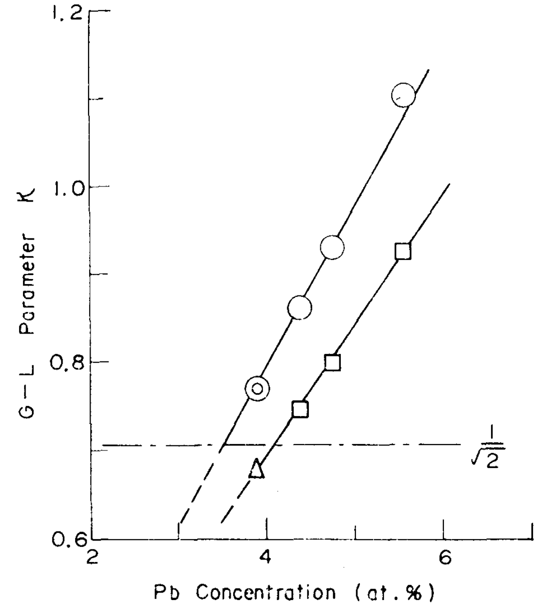

Fig. 11. Concentration dependence of G-L parameter in In-Pb alloy system. ◎ and △: $\kappa_1(0)$ and $\kappa_1(1)$, respectively, ○ and □: $\kappa_1(0)$ and $\kappa_1(1)$ in the previous work (I), respectively.

values are in good agreement with the results in I as shown in Fig. 11.

With the values of $\kappa_1(1)$ and $\kappa_1(0)$,

$$\frac{\kappa_1(0)}{\kappa_1(1)} = 1.13\,, \tag{9}$$

which is slightly smaller than the predicted value by Maki and again in good agreement with previous results (I). This trend of the concentration dependence of $\kappa_1(0)/\kappa_1(1)$ has already been described in I.

Thus, it is confirmed quantitatively that the In alloy containing 3.9 at. % Pb is a superconductor with $\kappa_1(1)=0.68$ and $\kappa_1(0)=0.77$ which shows type I to type II transition at $T_{cr}=2.2°\mathrm{K}$.

Summary

Type I to type II transition in the superconductivity of the sample studied is verified by measurements on the magnetization and the magnetic field dependence and the temperature dependence of the thermal conductivity. The critical temperature of the sample is determined to be 2.2°K.

The values of the residual resistivity $\rho_n=2.46\,\mu\Omega\mathrm{cm}$, the mean free path $l=5.69\times 10^{-6}\mathrm{cm}$, the transition temperature $T_c=3.65°\mathrm{K}$, and the G-L parameters $\kappa_1(0)=0.77$ and $\kappa_1(1)=0.68$ are in good agreement with the results in I. The ratio $\kappa_1(0)/\kappa_1(1)=1.13$ is slightly smaller than predicted value by Maki and is in good agreement with previous results (I).

Acknowledgements

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