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Author

Nembach, Eckhard.

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February 1968

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Eckhard Nembach

Inorganic Materials Research Division, Lawrence Radiation Laboratory,
University of California, Berkeley, California

ABSTRACT

T. Claeson's measurements of superconducting transition temperatures T_c of lead-thallium alloys have been repeated in the composition range 47-53 at.% Tl. In contrast to T. Claeson's findings no dip in the T_c versus composition-plot has been found. The exceptional behavior of T. Claeson's specimens with 49-50 at.% Tl seems to be due to sample inhomogeneity.

1. INTRODUCTION

The superconducting transition temperatures T_c of lead-thallium alloys have been measured over the whole composition range by T. Claeson.¹ His results are the following: adding up to about 80 at.% Tl decreases T_c monotonically from about 7.2°K to about 1.3°K, only specimens containing 49-50 at.% Tl showed an exceptional behavior. Their critical temperatures were 0.1 to 0.2°K lower than one would have expected by interpolating T_c 's of specimens with higher or lower thallium concentrations (see Fig. 1). Moreover, these specimens did show two different T_c 's. The author's explanation for this was as follows: Using data on the electron structure of pure lead, measured by J. R. Anderson and A. V. Gold² and applying the rigid band model, he finds that lead-thallium alloys with compositions of roughly 50 at.% Tl should have a very high density of electron states at the Fermi surface, because at the electron concentration of these alloys electrons just start entering the third band. T. Claeson suggested that the dip in the curve T_c versus composition is a result of this high density of electron states. Unfortunately he was not able to rule out the possibility of some phase change, such as ordering.

Therefore we did the same experiments for the system lead-indium, for which one expects a similar behavior. Because of the great difference in the atomic number it would be possible to detect ordering by x-ray diffraction. The results, which will be published elsewhere,³ were that there is no deviation of the plot T_c versus composition from a smooth curve. For this reason it was decided to repeat the measurements for the system lead-thallium.

2. SPECIMEN PREPARATION

The materials were provided by the American Smelting and Refining Company. According to the manufacturer the impurity content was less than 10 ppm. To avoid oxidation, the specimens were always handled in a dry-box under helium. The weighed amounts of the constituents were sealed under a reduced helium atmosphere in quartz bulbs, agitated at 440°C for 24 h in a rocking furnace, quenched in ice-water, cut to pieces (10-500 mm³) and sealed in cylindrical quartz-tubes, i.d. 6 mm, again under a reduced helium atmosphere. They were annealed in the same way as T. Claeson did it: At least 21 d at 295°C, followed by a quench in ice-water.

T_c has been measured in the apparatus described in reference (3), the absolute errors of T_c are estimated to be less than 15 mdeg, whereas relative errors will be smaller than 5 mdeg. In the described condition the specimens showed a large, reproducible hysteresis in the transition, T_c 's taken in cooling down or warming up differed up to 40 mdeg. Therefore the quartz-tubes were opened in a dry-box and sealed as before under helium. This reduced the average T_c hysteresis to 2 mdeg. The reason for this is probably that the helium left in the original quartz-tubes provided only a poor heat contact between specimen and thermometer. It has been proved that it was not the 40 min long exposure to room temperature during the change of the quartz tubes which reduced the hysteresis. It was not possible to melt and anneal the specimens in the same quartz-tube, as it had been done for the lead-indium specimens, because most of the quartz-tubes broke during the first quench.

3. RESULTS

The superconducting transition was detected by the change of the mutual inductance of two coaxial coils. If ΔL is the whole change in inductance, caused by the transition, then T_x^{NS} denotes the temperature at which the change in inductance was $\frac{x}{100} \Delta L$ for the normal to superconducting transition, and correspondingly T_x^{SN} , the temperature for the superconducting to normal transition. T_c was defined as $\left\{ \frac{T_{50}^{SN} + T_{50}^{NS}}{2} \right\}$. A measure for the transition width is Δ_{10} and Δ_{25} , which are defined as:

$$\Delta_{10} = (T_{50}^{SN} - T_{10}^{SN} + T_{10}^{NS} - T_{50}^{NS})$$

and

$$\Delta_{25} = (T_{50}^{SN} - T_{25}^{NS} + T_{25}^{NS} - T_{50}^{NS}).$$

In Table I, T_c , Δ_{10} , and Δ_{25} are listed for the various specimens tested. No second transition has been found within 0.8°K above or 1.0°K below the quoted T_c 's. Some specimens showed values for Δ_{10} as high as 200 mdeg. Therefore all specimens for which Δ_{10} exceeded 35 mdeg were melted and annealed once more. This changed T_c less than 10 mdeg but it decreased Δ_{10} markedly. In Fig. 1 our T_c 's are plotted together with those of T. Claeson. Within the absolute accuracy of 0.1°K of his temperature-measurements, our T_c 's and his coincide, except for specimens with thallium contents between 49 and 50 at.%, for which T. Claeson had found a deviation from a smooth curve. From our measurements it is clear that this special behavior does not exist. T. Claeson's findings are probably due to inhomogeneity of his specimens, because they were slowly cooled from the melt, which results in regions of various thallium contents in a sample. As the solidus and liquidus line in the lead-thallium phase diagram⁴ make only a rather

small angle with the concentration axis, cooling a specimen slowly through the two-phase region may bring about a great scatter in the composition, even though solidus- and liquidus-line are rather close together.

This view is supported from our experience that only fast quenching and, if necessary, remelting the specimens, resulted in a small transition width.

Therefore the conclusions drawn by T. Claeson from his measurements are no longer valid. Either the quoted high density of electron states, which T. Claeson suggested, does not exist or its influence on the superconducting transition temperature is counterbalanced by some other effect.

Acknowledgement - This work was supported by the United States Atomic Energy Commission.

Table I

Composition at. % Tl	Transition Temp. (°K)	Transition Width in 10^{-3} °K	
		$\Delta_{10} = T_{50}^{SN} - T_{10}^{SN} + T_{10}^{NS} - T_{50}^{NS}$	$\Delta_{25} = T_{50}^{SN} - T_{25}^{SN} + T_{10}^{NS} - T_{50}^{NS}$ *
47.42	5.637	27	2
48.80	5.572	4	1
49.14	5.557	4	1
49.16	5.554	2	0
49.33	5.547	6	3
49.60	5.534	4	2
49.81	5.521	4	2
50.05	5.512	7	1
50.30	5.497	13	1
50.40	5.491	15	3
50.54	5.483	27	2
50.61	5.481	12	4
50.71	5.477	7	2
50.91	5.464	18	3
51.17	5.452	6	3
51.71	5.422	4	1
52.55	5.374	11	0
53.60	5.312	34	3

* At the temperatures T_x^{SN} , T_x^{NS} x% of the change inductance had occurred during the superconducting to normal and the normal to superconducting transitions, respectively.

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1. T. Claeson, Phys. Rev. 147, 340 (1966).
2. J. R. Anderson, A. V. Gold, Phys. Rev. 139, A1459 (1965).
3. E. Nembach, J. Phys. Chem. Sol. (in press).
4. M. Hansen, K. Anderko, Constitution of Binary Alloys, (McGraw-Hill Book Company, New York, 1958), p. 1113.

FIGURE CAPTIONS

Fig. 1 Critical temperature as a function of the Tl-concentration

○ this investigation

I T. Claeson's transitions

| links the two transitions of a specimen

----- Interpolation of T. Claeson's T_c -values for Tl-contents
lower than 49 or higher than 51 at. %.

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