

**Physics.** — *Further investigations on the magnetic disturbance of the supraconducting state of alloys.* By W. J. DE HAAS and J. VOOGD.  
(Communication N<sup>o</sup>. 214b of the Physical Laboratory Leiden.)

(Communicated at the meeting of January 31, 1931).

§ 1. *Introduction.* In previous papers we have already discussed several measurements of the magnetic disturbance of the supraconducting state of alloys <sup>1)</sup>. We found as a general rule that per degree decrease of temperature the magnetic transition curve is shifted more towards the higher field intensities for the alloys than for the pure metals.

The most striking example of this is the eutectic of the system lead-bismuth. At 4<sup>o</sup>.2 K this alloy is still supraconductive in a magnetic field of 14000 Gauss and at 1<sup>o</sup>.9 K still in a field of 20.000 Gauss.

We continued these investigations with some other lead alloys and with the eutectic of the system gold-bismuth. These lead alloys were solid solutions of 15 % mercury in lead, of 40 % thallium in lead and of 35 % bismuth in lead.

§ 2. During the making of homogeneous solid solutions, the complication arises that the liquid mixture crystallizes inhomogeneously. This is because of the fact that at different stages of the crystallization the liquid mixture is in equilibrium with solid solutions of different constitutions. In the condensation nuclei therefore a solution of definite composition crystallizes and round them layers of solid solutions of different concentration are formed as the temperature is lowered. Generally these solid solutions are made homogeneous by slow congelation. Afterwards the mass is cleared from the still subsisting inhomogeneities in the following way. It is rolled and welded repeatedly so that it becomes very finely divided. Then it is heated during a long time just below the temperature of first formation of liquid. Then the last inhomogeneities must disappear by diffusion.

For our investigations this procedure has the great disadvantage, that the required mechanical processes are too dangerous for the purity of the metal.

That is why we worked out another method. We dripped namely very small drops of liquid of the right composition on a cooled metal block, where they are directly congealed. In this way a rod of the alloy is formed by the accumulated drops. Each of these congealed drops was inhomogeneous to the same degree and because of the small dimensions

---

<sup>1)</sup> Comm. Leiden N<sup>o</sup>. 199c, Comm. Leiden N<sup>o</sup>. 208b.

of the drops only a short time was required to make the rod homogeneous by diffusion.

The dripping of the liquid was effectuated in the following way. The metal of the required composition was contained in a thickwalled vertical glass tube *A* (fig. 1), which ended in a narrow opening of about 0.05 mm diameter. Round this tube *A* a second tube *B* was placed, which could be evacuated. At the height of the metal mixture and of the small orifice an electric resistance furnace (*C*) was placed round the surrounding tube and in the closed lower end of the latter a copper block *D* was placed, which was cooled by liquid air.

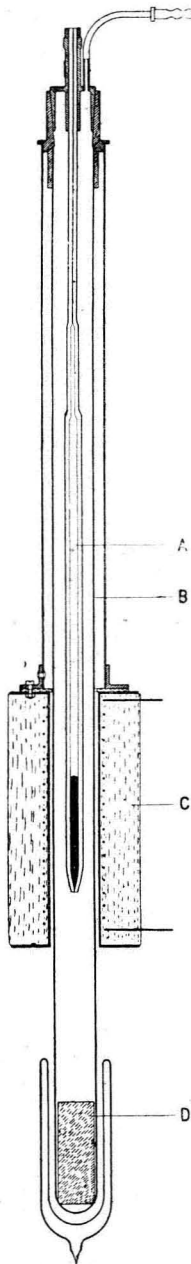


Fig. 1.

The metal mixture was melted and then the surrounding tube was evacuated. Now the liquid was pressed through the narrow orifice by means of carbonic dioxide of about 40 atm. Drops were formed of a few hundredths of a millimeter diameter, which were not solidified before they fell on the cold copper block or on the rod in formation.

We found an opening of about 0.05 mm to be very suitable for a pressure of 40 atm. A larger opening gives larger drops and so great a flux of metal, that the drops are not directly solidified on the block. With the same pressure a smaller opening makes the flux of the metal so weak, that at the opening the small drops are clustering together to large ones, which fall down as a whole.

For preliminary investigations with solid solutions we did use this wearisome procedure. The solid solution of 15 % mercury in lead and of 40 % thallium in mercury are made by cooling down very slow the liquid mixture.

The more extensive procedure has been used however for the preparation of solid solutions of bismuth in lead, both for those of 7 %, of 10 % and of 20 % bismuth in lead described in the preceding communication and for those of 35 % bismuth in lead the measurements with which will be discussed in this paper.

In the case of the latter alloy the rod was first made homogeneous by diffusion, then a wire was made by pressing it while cold; finally it was heated again during some time.

In order to control the method we made two resistances from different pieces of such a wire and determined for both the ratio of the

resistances at the boiling point of oxygen and at 20°. These ratios (about 0.7) were the same within 1 %.

§ 3. *Investigation of a solid solution of 35 % bismuth in lead.*

Of the system lead-bismuth we had investigated already the eutectic, in which the solid solution of bismuth in lead represented the only supraconductive phase.

For the percentage of bismuth in this solid solution, however, different values are given.

For our measurements it was necessary to know this percentage as accurately as possible. From the above mentioned investigations on solid solutions of 7 %, 10 % and 20 % bismuth in lead we knew that for a definite temperature the magnetic half value increases with the percentage of bismuth. For the solid solution with 20% bismuth the half value of the eutectic is not yet reached. Therefore the solid solution in the eutectic must have a higher percentage of bismuth than 20%. This is in agreement with some determinations of the melting point diagram of the system lead-bismuth which give for the percentage of the eutectic 35%.

That is why we made a homogeneous rod of the solid solution of 35% bismuth in lead with the procedure described above. For this rod we determined for different temperatures the transverse magnetic disturbance. The results are given in table 1.

Small deviations from the results of the measurements with the eutectic are found. These may be caused however by the complicated nature of the magnetic disturbance in the eutectic in which the current has to find its way through the layers of the supraconductive solid solution in the rod. This is namely the reason why the transverse disturbance cannot be measured purely for the eutectic, while for the homogeneous solid solution this is very well possible.

Still the quantitative agreement of the results is sufficient to allow the conclusion that the supraconductive solid solution in the eutectic contains about 35% bismuth.

§ 4. *Investigation of a solid solution of 40 % thallium in lead.*

Of the system thallium-lead we had already investigated the solid phase, the composition of which is represented by the formula  $PbTl_2$  and which becomes supraconducting at 4.09 K. Of a solution of 40% thallium in lead we expected the transition point to lie higher, so that in the helium region a considerable magnetic field would be required for the magnetic disturbance.

The rod was prepared by slow cooling and probably was still inhomogeneous. The results of the measurements are given in table 2.

The magnetical half value increases with about 1500 Gauss per degree decrease of temperature.

TABLE 1. *Pb-Bi* (35% *Bi*).

<i>H</i>	<i>R</i>	<i>T</i> ° K	<i>P</i> <sub>helium</sub> in mm Hg	<i>HW</i> <sup>1/2</sup>
14550	0	4.22	763	18450
15450	0.000081 Ω			
16750	136			
17800	516			
19200	1389			
20150	1675			
21150	1765			
22750	1809			
24200	1820			
26000	1822			
20150	1700			
17800	689			
17800	0	3.36	306	22050
19900	0.000075			
21150	421			
22100	963			
23300	1498			
25000	1748			
26000	1799			
27300	1817			
21150	472			
20450	0.000008	2.94	170	23450
22100	254			
24200	1311			
26000	1687			
27300	1783			
17800	0	2.43	68.3	24300
21150	0.000004			
22100	28			
24200	554			
26000	1388			
27300	1648			
22100	0.000002	1.97	22.9	25700
24200	234			
26000	1068			
27300	1459			
21150	0	1.88	16.9	26250
22100	0.000006			
23300	26			
25000	305			
26000	1325			

TABLE 2. *Pb-Tl* (40% Tl).

$H$	$R$	$T^\circ K$	$P_{helium}$ in mm Hg	$HW^{1/2}$
2300	0	4.23	765	3050
3050	0.000255 $\Omega$			
3800	447			
5300	500			
7500	524			
4500	0	2.93	168	5000
5300	0.000368			
6050	454			
7500	498			
11150	533			

§ 5. *Investigation of a solid solution of 15% mercury in lead.*

This solid solution was also prepared by slowly cooling down the liquid mixture. The results of the measurements are to be found in table 3. The

TABLE 3. *Pb-Hg* (15% Hg)

$H$	$R$	$T^\circ K$	$P_{helium}$ in mm Hg	$HW^{1/2}$
6500	0	4.23	765	6800
7500	0.000321 $\Omega$			
8250	548			
9000	579			
11150	612			
8950	0	2.93	168	10300
10450	0.000123			
11850	539			
13300	586			
14550	605			

magnetical half value increases with about 2700 Gauss per degree decrease of temperature.

§ 6. *Investigation of the eutectic of the system gold-bismuth.*

Finally we investigated the eutectic of the system gold-bismuth. For this purpose we used the resistance *Au-Bi II* the supraconducting state of which had been examined already in collaboration with EDM. VAN AUBEL <sup>1)</sup>.

The magnetic fields were parallel with the axis of the rod. They were generated by a solenoid placed round the helium cryostat. The results of the measurements are given in table 4. In the preceding cases we com-

TABLE 4. *Au-Bi II.*

<i>H</i>	<i>R</i>	<i>T</i> ° K	<i>P</i> <sub>helium</sub> in mm Hg	<i>HW</i> <sup>1/2</sup>
42.6	0.000050 Ω	1.54	5.0	95
63.8	481			
77.8	1020			
100.2	1963			
149.8	3134			
199.6	3612			
100.3	2021			
72.0	0	1.25	1.1	147
96.2	0.000273			
146.6	1786			
170.4	2454			
199.2	3041			
96.8	309			

pared the decrease of the magnetical half value with the temperature with that found for the pure supraconductive component of the alloy.

In the case of the eutectic however this is not possible, as it contains no supraconductive component.

The only thing we can do is to compare the decrease of the magnetic half value with that found for other pure supraconductors in the same temperature region e. g. for thallium.

We then find, that also an alloy of two non supraconductors shows a greater shift of the transition figure towards the high fields than a pure supraconducting metal.

<sup>1)</sup> Leiden Comm. 197a.

§ 7. *Summary.*

As result of our investigations we may give the rule : At a decrease of temperature the magnetic transition figure is more shifted towards the high fields for combinations than for pure metals. The validity of this rule has been proved for the:

- 1st solid solution of bismuth in tin,
- 2nd " " " cadmium " "
- 3rd " " " 7 % bismuth in lead,
- 4th " " " 10 % " " "
- 5th " " " 20 " " " "
- 6th " " " 35 " " " "
- 7th " " " 40 " thallium " "
- 8th " " " 15 " mercury " "
- 9th " " " 4 " bismuth " gold,
- 10th compound  $Bi_5Tl_3$ ,
- 11th "  $Sb_2Tl_7$ ,
- 12th "  $Sb_2Sn_3$ ,
- 13th "  $PbTl_2$ ,
- 14th eutectic bismuth-lead-tin,
- 15th " " " " -cadmium.

The highest field not disturbing the supraconductivity was observed for the solid solution of 35 % bismuth in lead at 1°.88 K and amounted to 21150 Gauss.

Finally we wish to express our thanks to Mr. H. BRUINING for his help during the measurements.

---