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A NEW COMPOUND IN Au-Pb ALLOY SYSTEM*

BY

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ABSTRACT

A new phase as observed by the electron diffraction method in the thin films of Au-Pb alloy was confirmed by both the metallographic and X-ray diffraction methods with bulk specimens. This new phase was an intermetallic compound whose composition was $AuPb_3$, and was formed peritectically like the compounds Au_2Pb and $AuPb_2$. The unit cell of $AuPb_3$ is tetragonal with lattice constants $a=11.958$ Å and $c=5.878$ Å. The measured density is 12.8 g cm^{-3} which agrees well with the calculated one on the basis of 8 molecules per unit cell (12.93 g cm^{-3}). The peritectic point of this phase is very close to the eutectic temperature (215°C).

1. Introduction

In Au-Pb alloy system, two intermetallic compounds, Au_2Pb and $AuPb_2$, have already been known. The former is cubic with lattice constant $a=7.93$ Å, while the latter is tetragonal with $a=7.31$ Å, $c=5.644$ Å, and both crystallize by the peritectic reaction.

By the electron diffraction study on Au-Pb alloy films prepared by the deposition of metal vapours in vacuum, the writer found a new phase (Fig. 1) in Pb-rich side of this alloy system, and its composition was estimated between the eutectic (15 wt. % Au) and $AuPb_2$ (32.2 wt. % Au). This new phase seemed to be stable but not metastable one. In order to confirm this and to determine the accurate composition and the lattice dimensions, the writer carried out the X-ray diffraction and metallographic investigations with bulk specimens.

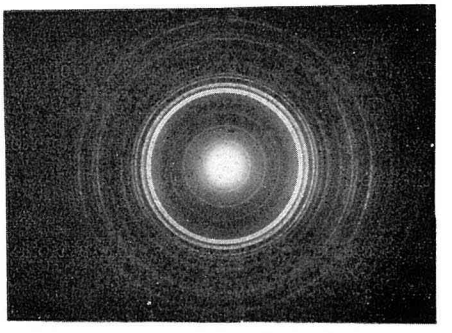

Fig. 1. The electron diffraction pattern of the new phase.

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2. Experimental procedure

Specimens used are 99.99% Au and Merck's Pb. Specimens were vacuum sealed in Terex glass tubes (6~8 mm in diameter and about 1 mm in wall thickness). The amount of specimen in each tube was about 10 g. These specimens were melted in a salt bath at about 400°C for 15~30 minutes, quenched in a mixture of ice and NaCl or air-cooled to room temperatures, and then annealed for a suitable time at about 200°C. After these heat treatments, specimens were polished by emery papers, tooth-paste and silk velvet, etched in the solution of 5% nitric acid in absolute alcohol and then investigated microscopically.

A part of each especimen used in the microscopical study was examined by the X-ray diffraction with powder method using a Cu K_α radiation with Ni filter.

3. Experimental results

(a) *Determination of composition*

Fig. 2 shows a photomicrograph of the typical eutectic structure obtained by the air-cooled specimen of an alloy containing 15 wt. % Au. This specimen does not show any appreciable change of texture after annealing for about 24 hours at about 200°C. An X-ray powder pattern obtained by this specimen showed the presence of Pb and the new phase, thus indicating that the latter is surely present in bulk specimens and its composition is between $AuPb_2$ and the eutectic. Then, starting from the eutectic composition, specimens differing by one or two wt. % Au were prepared and examined microscopically as well as by X-ray diffraction method after heat treatment.

Specimens in the range of compositions of 17~23 wt. % Au indicate two phases of the eutectic and the new phase, one of the photomicrographs being shown in Fig. 3. From the facts that the X-ray patterns of these specimens show very intense diffraction rings of the new phase, it is known that the large grains appeared in Fig. 3 are the crystals of the new phase.

The specimens containing 24 wt. % Au show one phase after suitable annealing, as is shown in Fig. 4. X-ray pattern of this annealed specimen also shows that of the new phase only (Fig. 9).

The alloy containing 25 wt. % Au shows two phases after annealing (see Fig. 5). It is confirmed by X-ray diffraction that these two phases are the new phase and $AuPb_2$.

From the results mentioned above, it seems that this new phase has no appreciable homogeneous range but is a compound like Au_2Pb or $AuPb_2$, and this result should be compared with a calculated value of 24.08 wt. % Au for the formula unit $AuPb_3$.

(b) *Crystal system and unit cell dimensions*

The X-ray diffraction pattern of this new compound (Fig. 9) can be indexed on

Table I.

(*hkl*)	*d* (calc.)	*d* (obs.)	*I* (obs.)
(110)	8.456Å	(8.46Å)*	w
(200)	5.979	(5.92)*	vw
(101)	5.275	(5.29)*	w
(220)	4.224	(4.22)*	vw
(211)	3.956	(3.96)*	w
(310)	3.781	(3.78)*	vw
(301)	3.299	(3.30)*	vw
(002)	2.939	(2.98)*	vvw
(321)	2.888	2.890	s
(330)	2.818	2.822	vs
(112)	2.776	2.781	m
(420)	2.674	2.672	w
(411)	2.600	2.597	vs
(222)	2.413	2.418	s
(510)	2.345	2.338	m
(312)	2.321	2.314	m
(520)	2.221	2.227	vw
(402)	2.096	2.098	vvw
(521)	2.077	2.075	vw
(611)	1.864	1.863	m
(710)	1.691	1.691	s
(602)	1.650	1.651	w
(711)	1.625	1.628	s
(721)	1.582	1.584	w
(542)	1.576	1.574	vw
(650)	1.531	1.533	vw
(731)	1.517	1.519	vw
(004, 523)	1.469	1.470	s
(741, 811)	1.438	1.438	m
(660)	1.409	1.408	vvw
(821)	1.408		
(613, 224)	1.388	1.388	w
(831)	1.362	1.361	w
(840)	1.337	1.335	vw
(910)	1.321	1.320	vvw
(414)	1.311	1.307	w
(901)	1.296	1.293	m
(921, 761)	1.267	1.265	vw
(504, 434)	1.252	1.249	vvw
(851)	1.239	1.240	vw
(444, 653)	1.206	1.205	w
(771)	1.184	1.183	w
(852, 115)	1.164	1.164	vvw
(951)	1.140	1.139	vw
(704)	1.112	1.114	w
(724, 913)	1.095	1.095	w
(844, 10.33)	0.988	0.988_8	vvw
(981, 12.11)	0.977	0.979_2	vvw
(854)	0.959	0.959_8	vvw
(226, 12.31)	0.953	0.954_0	vvw
(406)	0.931	0.930_9	vvw
(805)	0.923	0.924_0	vvw

* These lines were observed by the electron diffraction method.

the basis of a tetragonal unit cell with $a=11.958$ Å and $c=5.878$ Å. The observed and calculated spacings are shown in Table I. (The X-ray diffraction rings corresponding to large spacings were difficult to measure, and therefore the values of these spacings were taken from the electron diffraction data.)

The density of $AuPb_3$ measured by Yoshida and Takei's method (**1**) was found to be 12.8 g cm^{-3} which agrees well with the value 12.93 g cm^{-3} calculated on the basis that the unit cell contains 8 molecules.

(c) *Transformation by the peritectic reaction*

Fig. 6 shows a microphotograph of quenched specimen containing 24 wt. % Au. This shows only two phases. X-ray pattern of this specimen shows the diffraction rings of $AuPb_2$, $AuPb_3$ and Pb, and the relative intensity of $AuPb_3$ and Pb is similar to that of eutectic. This shows that these two phases are $AuPb_2$ and eutectic, but not $AuPb_3$ phase. Similar results were obtained for air-cooled specimens of other compositions between 26 wt. % Au and 17 wt. % Au.

The quenched specimen of 24 wt. % Au transforms easily into one phase after the annealing for about 26.5 hours at about 200°C (Fig. 4), but the air-cooled one of the same composition (Fig. 7) does not completely transform into one phase even after the prolonged annealing for about 60 hours. (see Fig. 8). These facts and the results mentioned in section (a) show that the $AuPb_3$ phase is formed by the peritectic reaction.

The incompletion of transformation in air-cooled specimens may depend upon the fact that the grain size of $AuPb_2$ is relatively large compared with the grain size of

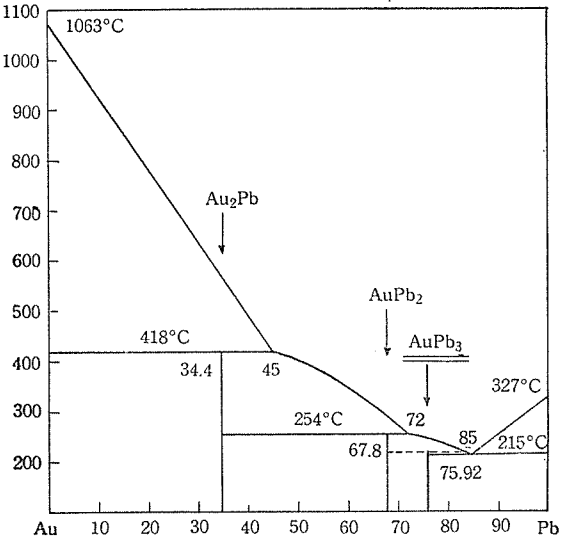

Fig. 10. Phase diagram of Au-Pb alloy system. The position of $AuPb_3$ is shown.

$AuPb_2$ in quenched specimens, and the rate of peritectic reaction is almost interrupted after these grains were surrounded by the new phase.

The fact that the $AuPb_2$ phase presents itself even in the quenched or air-cooled specimens of 17 wt. % Au and the $AuPb_3$ phase is absent suggests that the peritectic point of $AuPb_3$ is very close to the eutectic temperature. In fact, the electron diffraction observations on the highest temperature of stability for the $AuPb_3$ phase support this suggestion (Fig. 10), but the precise determination of the peritectic point requires further investigations.

4. Summary

In Au-Pb alloy system, a new compound was found, the composition of which was metallographically determined to be $AuPb_3$. It was determined by X-ray diffraction method that the unit cell of this compound is tetragonal with lattice constants $a=11.958$ Å and $c=5.878$ Å containing 8 molecules. The measured density was 12.8 g cm^{-3}, while the calculated one was 12.93 g cm^{-3}. It was also confirmed by the metallographic study that the new compound $AuPb_3$ is formed peritectically like Au_2Pb or $AuPb_2$ and the peritectic point of this compound is very close to the eutectic temperature (215°C).

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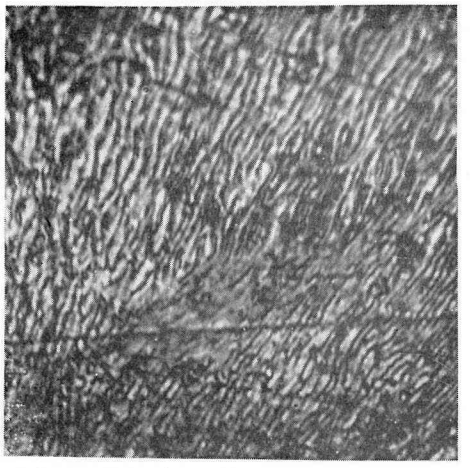

Fig. 2. ×800
15 wt. % Au, air-cooled
(eutectic)

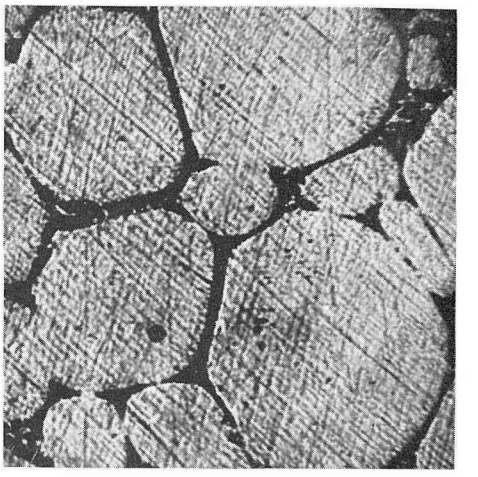

Fig. 3. ×150
23 wt. % Au, annealed for 28 hours at 200°C
($AuPb_3$+eutectic)

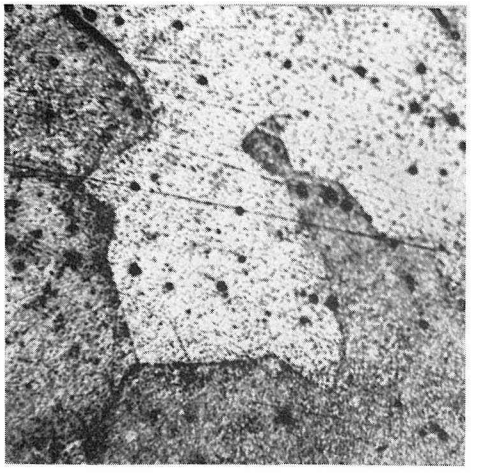

Fig. 4. ×150
24 wt. % Au, quenched and annealed for 26.5 hours at 200°C
($AuPb_3$)

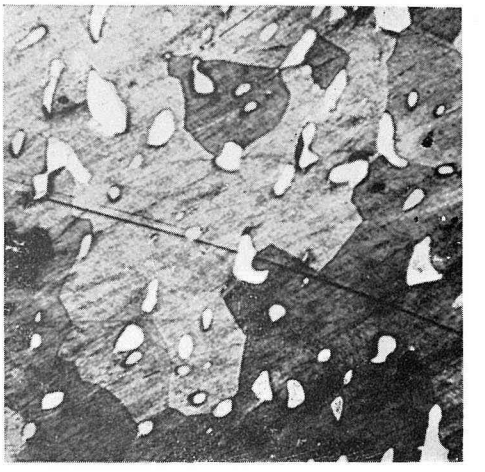

Fig. 5. ×150
25 wt. % Au, annealed for 53 hours at 200°C
($AuPb_3$+$AuPb_2$)

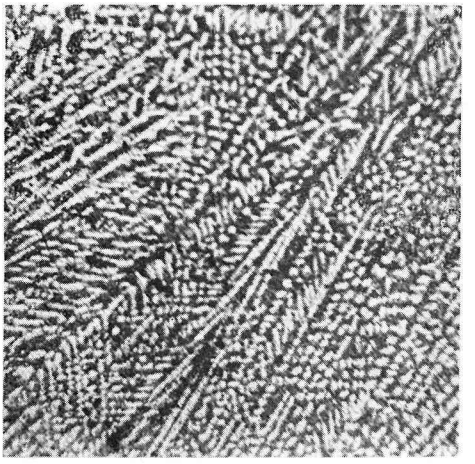

Fig. 6. ×150

24 wt. % Au, quenched
($AuPb_2$+eutectic)

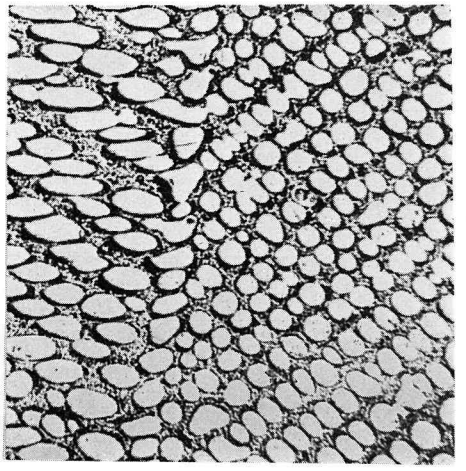

Fig. 7. ×150

24 wt. % Au, air-cooled
($AuPb_2$+eutectic)

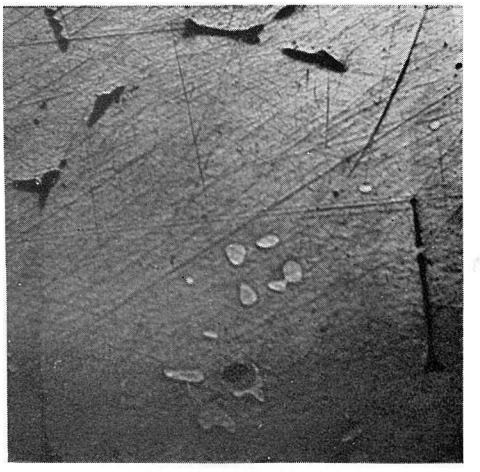

Fig. 8. ×150

24 wt. % Au, air-cooled and annealed for 60 hours at 200°C
($AuPb_3$+$AuPb_2$+eutectic)

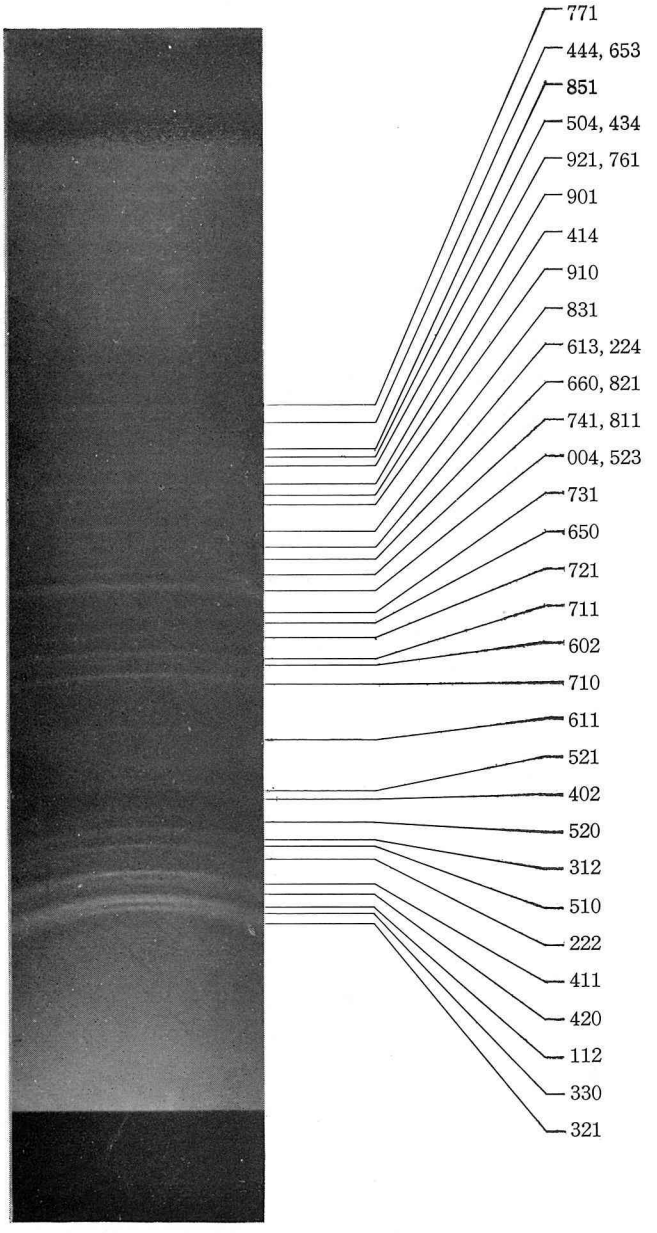

Fig. 9.

24 wt. % Au–Pb ($AuPb_3$)
Tetragonal $a_0 = 11.95_8$Å
$c_0 = 5.87_8$A