1970 (Academic Press of Japan, Tokyo, 1970), Vol. 12, p. 361.
[14]E. I. Katz, Zh. Eksperim. i Teor. Fiz. 56, 1675 (1969) [Sov. Phys. JETP 29, 897 (1969)].
[15]E. I. Katz, Zh. Eksperim. i Teor. Fiz. 58, 1471 (1970) [Sov. Phys. JETP 31, 787 (1970)].
[16]F. R. Gamble, F. J. Di Salva, R. A. Klemm, and T. H. Geballe, Science 168, 568 (1970).
[17]F. J. Di Salva, R. Schwall, T. H. Geballe, F. R. Gamble, and J. H. Osiecki, Phys. Rev. Letters 27, 310 (1971).
[18]T. H. Geballe, A. Menth, F. J. Di Salva, and F. R. Gamble, Phys. Rev. Letters 27, 314 (1971).

PHYSICAL REVIEW B VOLUME 5, NUMBER 3 1 FEBRUARY 1972

Occurrence of Superconductivity in Simple-Cubic $(Au_{1-x}Pd_x)Te_2$ Alloys

Wilkie Y. K. Chen and C. C. Tsuei
W. M. Keck Laboratory of Engineering Materials,
California Institute of Technology, Pasadena, California 91109
(Received 23 September 1971)

The superconducting transition temperatures and lattice parameters of metastable simple-cubic alloys $(Au_{1-x}Pd_x)Te_2$, with $0 \leq x \leq 0.6$, obtained by rapid quenching from liquid state, have been measured as a function of x. As x increases the transition temperature T_c goes through a minimum of 1.6 °K near $x = 0.05$, and then increases steadily to a maximum of 4.5 °K at $x = 0.45$. The lattice parameter shows a slight linear decrease as x increases. The transverse magnetoresistance for the composition $x = 0.2$ saturates at a relatively low field (~2 kG) and is unusually large (~0.14). The initial depression of T_c with increasing Pd content can be interpreted in terms of the information obtained in a previous study of binary Au-Te alloys. The unusual increase of T_c with further increase in the Pd concentration is not well understood.

I. INTRODUCTION

Superconductivity in metastable simple-cubic alloys (with one atom per unit cell) has been reported previously.[1-5] It was also pointed out that all the simple-cubic alloy phases found so far are superconductors with transition temperatures ranging from approximately 1 to 7 °K.[4] These findings suggest that this rather unusual crystal structure is favorable for the occurrence of superconductivity. Among the simple-cubic alloys, the Au-Te system has been studied in some detail.[5,6] It has been shown that the anomalies in the variation of lattice parameter, thermoelectric power, and superconducting transition temperature (T_c) with concentration in this alloy system can be qualitatively explained in terms of a Fermi-surface–Brillouin-zone interaction. A study of the effect of magnetic-impurity atoms such as Mn and Fe on the lattice parameter and on the superconducting transition temperature of the Au-Te alloys has been published.[6] The results of this study give additional support to the electronic band structure proposed for the simple-cubic Au-Te alloys.[5] Furthermore, the results also show that the addition of Fe or Mn monotonically decreases the superconducting transition temperature. The rate of depression of T_c by Fe, however, is about seven times smaller than that by Mn. This is consistent with the fact that Mn carries a localized magnetic moment in the Au-Te alloys while Fe does not.

A preliminary study has shown that Pd can be substituted for Au in $AuTe_2$ alloys, and these alloys are superconducting.[4] In view of the fact that the element Pd is nearly ferromagnetic it seems to be worthwhile to study the compositional dependence of T_c and the lattice parameter as the Au in $AuTe_2$ is replaced by Pd.

II. EXPERIMENTAL PROCEDURE

The compositions of the alloys investigated are of the form $(Au_{1-x}Pd_x)Te_2$, where $0 \leq x \leq 0.6$. The alloys were prepared by induction melting of the appropriate quantities of the constituents (99.999% pure Te and 99.99% pure Au, Pd) in quartz crucibles under an argon atmosphere. The weight losses were found to be less than 0.2% so the nominal compositions of the alloys were taken as the actual ones. The simple-cubic phase was obtained by liquid quenching at a rate of about 10^7 °C/sec, using the "gun" technique.[7] The structure of each quenched specimen was checked by taking diffraction patterns with a Norelco x-ray diffractometer (Cu $K\alpha$ radiation). The lattice parameters of the simple-cubic phase were obtained from Debye-Scherrer films using the Nelson-Riley extrapolation function. The results of the lattice-parameter measurements are shown in Fig. 1. It was found that it became increasingly difficult to obtain a single phase for alloys with x approaching 0.5. In the x-ray dif-

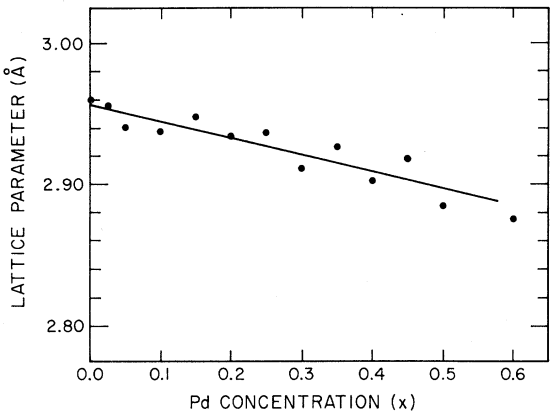

FIG. 1. Lattice parameter of simple-cubic alloys $(Au_{1-x}Pd_x)Te_2$.

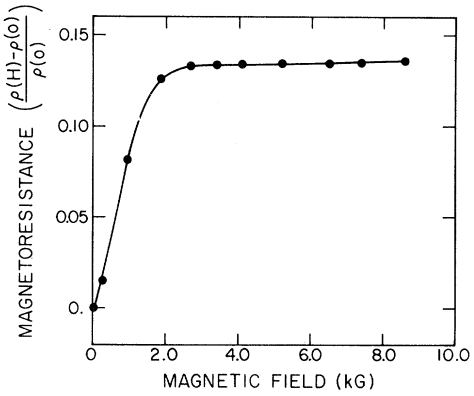

FIG. 3. Transverse magnetoresistance of the simple-cubic alloy $(Au_{80}Pd_{20})Te_2$ measured at 4.2 °K.

fraction pattern very faint lines from the second phase were detected and they were identified as those of $PdTe_2$ (ASTM x-ray data cards). In view of the very weak line intensity of the $PdTe_2$ phase, the fact that it is not superconducting at the T_c of all the simple-cubic phase studied in this work,[8] and the fact that the lattice parameter of the simple-cubic phase decreases linearly with increasing Pd concentration (Fig. 1), the presence of a small amount of $PdTe_2$ in some samples should not affect the result of the T_c measurement in any significant manner. The superconducting transition temperature was obtained by a standard ac bridge technique, using a frequency of 1 kHz. The magnetic field at the sample location was estimated as a few gauss. The temperature corresponding to the onset of transition was taken to be the transition temperature. The temperature was measured with a Honeywell germanium resistance thermometer calibrated against the 1958 He^4 vapor-pressure scale of temperature.[9]

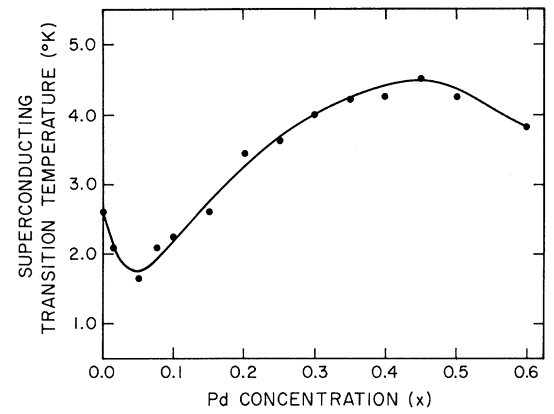

FIG. 2. Superconducting transition temperature (T_c) of the simple-cubic alloys $(Au_{1-x}Pd_x)Te_2$.

Magnetic-susceptibility data would be very useful in analyzing the alloying effect on the superconducting transition temperature of the $(Au_{1-x}Pd_x)Te_2$ alloys. As explained in Ref. 6, a direct measurement of magnetic susceptibility at low temperature with the available magnetometer would have required at least 100 rapidly quenched samples. For this reason, such measurements were considered as impractical and transverse magnetoresistances were measured instead, to gain some insight as to the nature of the magnetic states of the Pd-Au-Te alloys. The experimental details were described in Refs. 5 and 6.

III. RESULTS AND DISCUSSION

As mentioned in Sec. II, the lattice parameters of the simple-cubic alloys decrease linearly with increasing Pd content as shown in Fig. 1. This observation is consistent with the notion that the atomic radius of Pd is smaller than that of Au. The results of the superconducting transition-temperature measurement are presented in Fig. 2. As Pd replaces Au atoms in the simple-cubic $AuTe_2$, the transition temperature decreases initially and then increases with Pd content for alloys $0.05 < x < 0.45$. For alloys with $x > 0.45$, the transition temperature decreases slightly with increasing Pd concentration. The observed effect of Pd on the superconducting transition temperature is quite interesting since both Fe and Mn monotonically decrease the T_c of the simple-cubic alloys.[6] The results of the magnetoresistance measurement at 4.2 °K shown in Fig. 3 for the alloy $Au_{0.8}Pd_{0.2}Te_2$ indicate a large and positive magnetoresistance which saturates at a magnetic field of only 2.5 kG. It should be mentioned that the transverse magnetoresistance of the simple-cubic Au-Te alloys containing Fe or Mn is of the order of 0.01%, whereas that of the $Au_{0.8}Pd_{0.2}Te_2$ alloy is 10%.

Positive magnetoresistance suggests the absence of localized moments in these alloys. The observed large magnetoresistance (comparable with that of Pd) could mean that the exchange-enhancement effect prevails in the alloys studied.

To further understand the T_c data, a brief discussion on the possible effect of Pd on the energy band and Fermi energy of $AuTe_2$ is in order. One possibility is that in analogy with the Au-Pd alloys, the Fermi energy decreases as Pd replaces Au and the assumption of a rigid band holds for alloys with $x < 0.05$. If this is the case, the Fermi surface will be farther away from the Brillouin zone with increasing Pd concentration. The fact that the magnetoresistance saturates at relatively low fields suggests that the conduction electrons at the Fermi surface execute closed orbits in the presence of external magnetic field. This is certainly consistent with the idea that the Fermi surface in Au-Pd-Te alloys does not contact the Brillouin zone.[5] Based on the information obtained from studying binary simple-cubic Au-Te alloys, a lowering of the Fermi level of the $AuTe_2$ alloy has the effect of decreasing the T_c.[5] The impurity effect itself, as shown in the simple-cubic Au-Te solutions containing Fe or Mn,[6] also tends to lower the superconducting transition temperature. The initial decrease of T_c with increasing Pd (Fig. 2) probably can be attributed to the above-mentioned effects. The reason for the further increase of T_c with Pd content is not obvious at the present time. The results of this investigation, however, bear a close resemblance to those reported by Hill and Matthias[10] on the T_c of the superconducting compounds U_6X (X = Mn, Fe, Co, and Ni) as a function of the average number of valence electrons per atom of the X component, In that case, a correlation is found between the superconducting transition temperature and the saturation magnetization of the X elements. The exact nature and the importance of the interaction between the $5f$ and $6d$ electrons of U and the $3d$ electrons of X in relationship to superconductivity is not known. The results of the present study seem to be less ambiguous since the basic alloy $AuTe_2$ is an s-p superconductor. The d electrons of Pd probably are responsible for the maximum observed in the T_c-vs-x plot as shown in Fig. 2. It might be worthwhile to mention that the oscillatory behavior of T_c as a function of transition-element concentration (Pd in Au-Te alloys, or X in U_6X compounds) could be related to the well-known Matthias's rule.

In summary, we have observed a minimum and a maximum in the variation of T_c with Pd content (x) in the simple-cubic alloys $Au_{1-x}Pd_xTe_2$. The initial depression of T_c with increasing Pd content can be interpreted in terms of the information obtained in the study of binary Au-Te alloys. The unusual increase of T_c as the Pd concentration is further increased is now well understood, but might be due to the effect of the $3d$ electrons in Pd.

ACKNOWLEDGMENT

The authors wish to express their appreciation to Professor Pol Duwez for his advice and encouragement throughout this work.

*Work supported by the U. S. Atomic Energy Commission.

[1]H. L. Luo, M. F. Merriam, and D. C. Hamilton, Science 145, 581 (1964).

[2]H. E. Bommel, A. J. Dornell, W. F. Libby, B. R. Tittmann, and A. J. Yencha, Science 141, 714 (1963).

[3]J. Wittig and B. T. Matthias, Science 160, 994 (1968).

[4]C. C. Tsuei, H. C. Yen, and P. Duwez, Phys. Letters 34A, 80 (1971).

[5]C. C. Tsuei and L. R. Newkirk, Phys. Rev. 183, 619 (1969).

[6]L. R. Newkirk and C. C. Tsuei, Phys. Rev. B 3, 735 (1971).

[7]P. Duwez and R. H. Willens, Trans. AIME 227, 362 (1962).

[8]C. J. Raub, V. B. Compton, T. H. Geballe, B. T. Matthias, J. P. Moita, and G. W. Hull, Jr., J. Phys. Chem. Solids 26, 2051 (1965). Furthermore, after rapid quenching, the T_c of equilibrium $PdTe_2$ drops from the reported value of 1.69 to 1.4 °K.

[9]H. von Dijk, M. Durieux, J. R. Clement, and J. K. Logan, Natl. Bur. Std. (U. S.) Monograph No. 10 (1960), part 2.

[10]H. H. Hill and B. T. Matthias, Phys. Rev. 168, 464 (1968).