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THE OCCURRENCE OF  
SUPERCONDUCTIVITY IN  
SIMPLE CUBIC  
(Au<sub>1-x</sub>Pd<sub>x</sub>)Te<sub>2</sub> ALLOYS

MASTER

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## ABSTRACT

The superconducting transition temperatures and lattice parameters of metastable simple cubic alloys  $(\text{Au}_{1-x}\text{Pd}_x)\text{Te}_2$ , 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  $1.6^\circ\text{K}$  near  $x = 0.05$ , and then increases steadily to a maximum of  $4.5^\circ\text{K}$  at  $x = 0.45$ . The lattice parameter shows a slight linear decrease as  $x$  increases. Transverse magnetoresistance for the composition  $x = 0.2$  saturates at relatively low field ( $\sim 2$  kG) and is unusually large ( $\sim 0.14$ ).

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 Pd concentration is not well understood.

## I. INTRODUCTION

Superconductivity in metastable simple cubic alloys (with one atom per unit cell) has been reported previously.<sup>1-5</sup> 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.<sup>4</sup> These findings suggest that this rather unusual crystal structure is favorable for the occurrence of superconductivity. Among the simple cubic alloys, Au-Te system has been studied in some detail.<sup>5,6</sup> 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 lattice parameter and on superconducting transition temperature of the Au-Te alloys has been published.<sup>6</sup> The results of this study gives additional support for the electronic band structure proposed for the simple-cubic Au-Te alloys.<sup>5</sup> 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 Au Te<sub>2</sub> alloys, and these alloys are superconducting.<sup>4</sup> 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 lattice parameter as the Au

in Au Te<sub>2</sub> is replaced in Pd.

## II. EXPERIMENTAL PROCEDURE

The compositions of the alloys investigated are of the form of Au<sub>1-x</sub>Pd<sub>x</sub>Te<sub>2</sub> 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 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.<sup>7</sup> The structure of each quenched specimen was checked by taking diffraction patterns with a Norelco x-ray diffractometer (Cu K $\alpha$  radiation). The lattice parameter 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 diffraction pattern very faint lines from the second phase were detected and were identified as those of Pd Te<sub>2</sub> (ASTM x-ray data cards). In view of the very weak line intensity of the Pd Te<sub>2</sub> phase, the fact that it is not superconducting at the T<sub>c</sub> of all the simple cubic phase studied in this work,<sup>10</sup> and the fact that the lattice parameter of simple cubic phase decreases linearly with increasing Pd concentration (Fig. 1) the presence of a small amount of Pd Te<sub>2</sub> in some samples should not effect the result of T<sub>c</sub> measurement in any significant manner. The superconducting transition temperature was obtained by

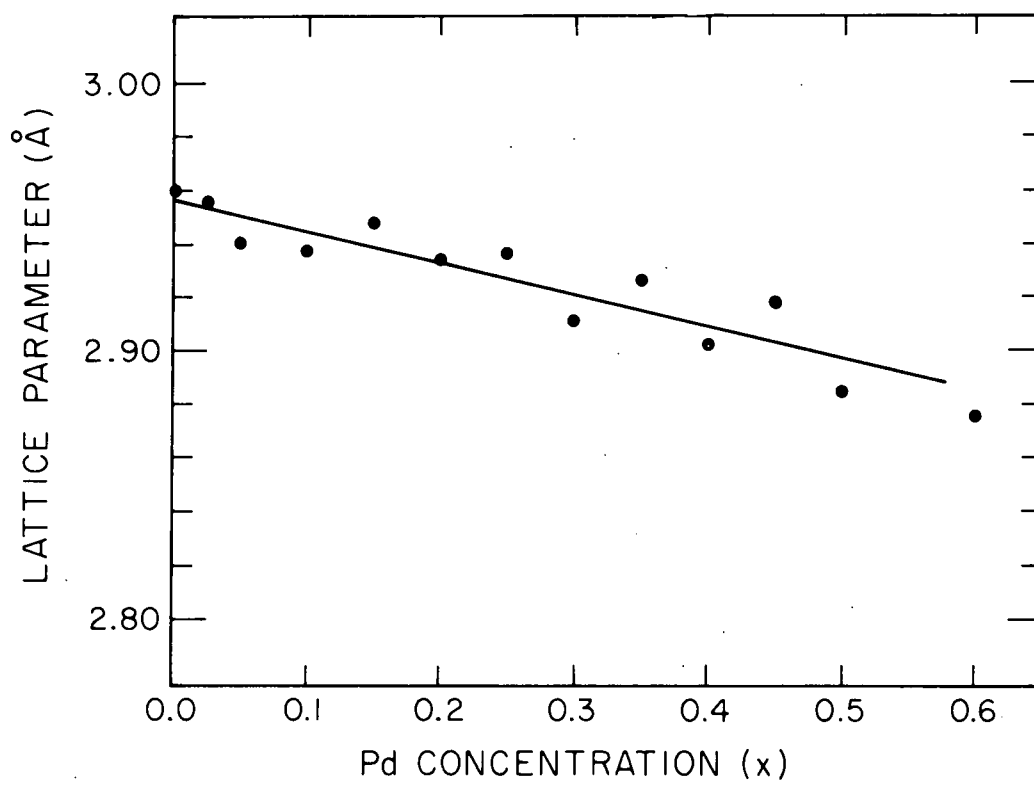


Fig. 1. Lattice parameter of simple cubic alloys  $(\text{Au}_{1-x}\text{Pd}_x)\text{Te}_2$ .

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<sup>4</sup> vapor pressure scale of temperature.<sup>9</sup>

Magnetic susceptibility data would be very useful in analyzing the alloying effect on the superconducting transition temperature of the (Au<sub>1-x</sub>Pd<sub>x</sub>)Te<sub>2</sub> alloys. As explained in Ref. 6, a direct measurement of magnetic susceptibility at low temperature with the available magnetometer would have required at least one hundred rapidly-quenched samples. For this reason, such measurements were considered as impractical and transverse magnetoresistance 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 Ref. 5, 6.

### III. RESULTS AND DISCUSSION

As mentioned in the previous section, 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 present in Fig. 2. As Pd replaces Au atoms in the simple cubic AuTe<sub>2</sub>, the transition temperature decreases initially and then increases with Pd content for alloys 0.05 < x < 0.45.

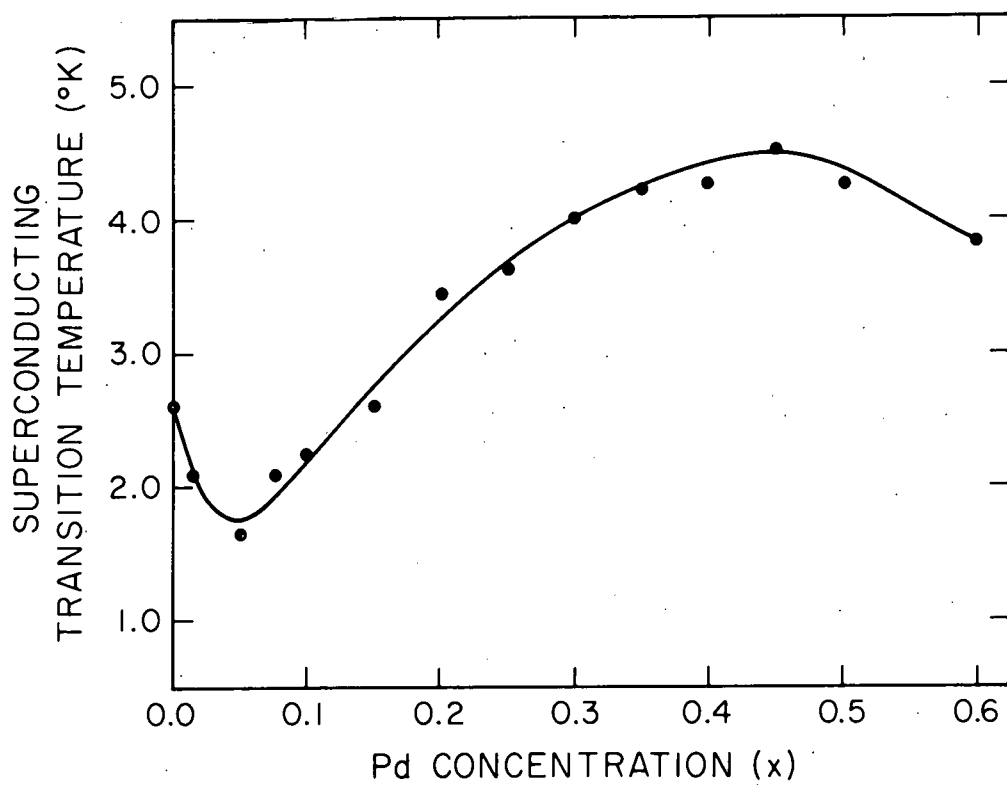


Fig. 2. Superconducting transition temperature ( $T_c$ ) of the simple cubic alloys  $(Au_{1-x}Pd_x)Te_2$ .

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.<sup>6</sup> The results of magnetoresistance measurement at 4.2°K shown in Fig. 3 for the alloy  $\text{Au}_{0.8}\text{Pd}_{0.2}\text{Te}_2$  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 Te-Au alloys containing Fe or Mn is of the order of 0.01% whereas that of the  $\text{Au}_{0.8}\text{Pd}_{0.2}\text{Te}_2$  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  $\text{Au Te}_2$  is in order. One possibility is that in analogy with 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.<sup>5</sup> Based on the information obtained from studying binary simple cubic Au-Te alloys, a lowering of the Fermi level of the  $\text{Au Te}_2$  alloy has the effect of decreasing the  $T_c$ .<sup>5</sup> The impurity effect itself as

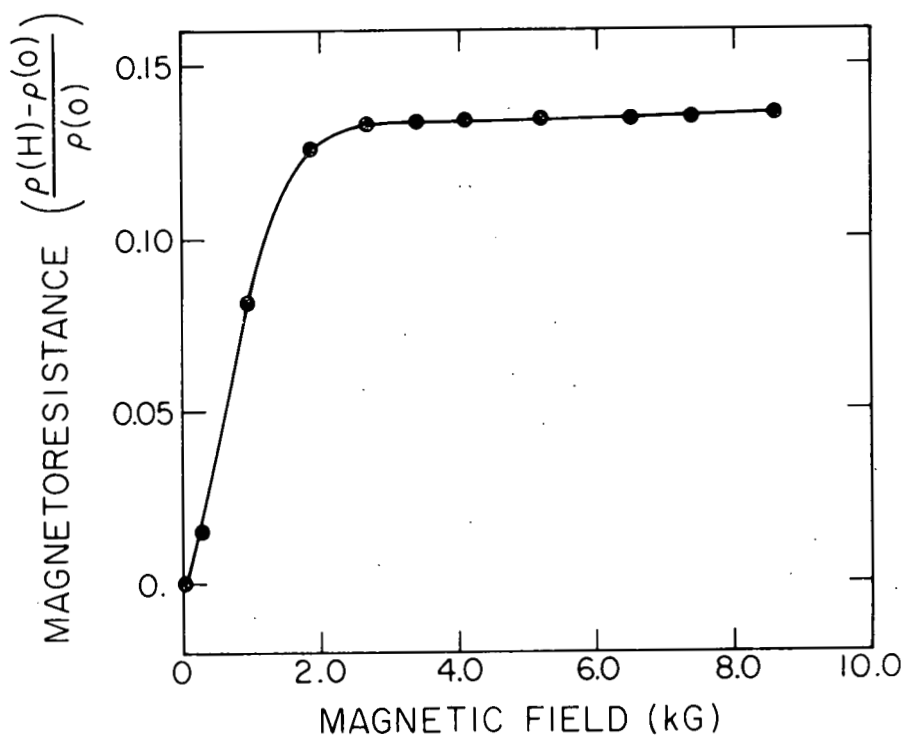


Fig. 3. Transverse magnetoresistance of the simple cubic alloy  $(\text{Au}_{80}\text{Pd}_{20})\text{Te}_2$  measured at  $4.2^\circ\text{K}$ .

shown in the simple cubic Au-Te solutions containing Fe or Mn<sup>6</sup> 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<sup>8</sup> on the  $T_c$  of the superconducting compounds  $U_6X$  ( $X = \text{Mn, Fe, Co and Ni}$ ) 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 5f, 6d electrons of U and the 3d electrons of X in relationship with superconductivity is not known. The results of the present study seem to be less ambiguous since the basic alloy  $\text{AuTe}_2$  is a s-d 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  $\text{Au}_{1-x}\text{Pd}_x\text{Te}_2$ . 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 not well understood, but might be due to the effect of the 4d electrons in Pd.

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Furthermore, after rapid quenching, the  $T_c$  of equilibrium Pd Te<sub>2</sub> drops from the reported value of 1.69°K to 1.4°K.