

It is easy to verify that these response functions satisfy the key identity⁶ which ensured gauge-invariant results,

$$\begin{aligned} & \left(\tilde{\chi}_{jj}(\vec{k}, -\vec{p}, \omega) - \frac{n_0(\vec{k}-\vec{p})}{m} \right) \vec{p} \\ &= \omega \tilde{\chi}_{jn}(\vec{k}, -\vec{p}, \omega) \quad , \end{aligned} \quad (\text{A20})$$

where we recall that in a homogeneous system $n_0(\vec{k}) = n_0 \delta(\vec{k})$.

Using a procedure analogous to that carried out in Ref. 4, we expand the RPA screened correlation functions in (A14) in powers of $\vec{k} \cdot \vec{p}/\omega$. Making use of

$$\int \frac{d\vec{p}}{(2\pi)^3} \frac{\vec{p}}{m} f_0(\vec{p}, \vec{k}) = 0, \quad \int \frac{d\vec{p}}{(2\pi)^3} f_0(\vec{p}, \vec{k}) = n_0(\vec{k}) \quad , \quad (\text{A21})$$

we find

$$\begin{aligned} \delta_{jP}^{\vec{k}}(\vec{k}, \omega) &= e^2 \int \frac{d\vec{p}}{(2\pi)^3} \left[\frac{1}{\omega} \left(\frac{\vec{p}}{m} \right) n_0(\vec{k}-\vec{p}) + O\left(\frac{1}{\omega^3}\right) \right] \\ &\times \delta\phi^{\text{eff}}(\vec{p}, \omega) + O\left(\frac{1}{\omega^2} \delta\vec{A}^{\text{eff}}\right) . \end{aligned} \quad (\text{A22})$$

We note that these lowest-order results imply

$$\begin{aligned} \tilde{\chi}_{jn}(\vec{k}, -\vec{p}, \omega) &= -(e^2/m) (\vec{p}/\omega) n_0(\vec{k}-\vec{p}) + O(1/\omega^3) \\ \tilde{\chi}_{jj}(\vec{k}, -\vec{p}, \omega) &= O(1/\omega^2) , \end{aligned} \quad (\text{A23})$$

which satisfy (A20) and hence are gauge invariant to order ω^{-1} . While we have not written down the explicit expression for $\tilde{\chi}_{jj}$ to order ω^{-2} , we emphasize that it is quite simple to obtain by using the first iteration of (A9) with

$$\left(\omega - \frac{\vec{p} \cdot \vec{k}}{m} \right)^{-1} = \frac{1}{\omega} + \frac{\vec{p} \cdot \vec{k}}{m\omega^2} + \dots \quad (\text{A24})$$

Using (A23) in (2.11) we arrive at (1.5), which is the basis of the discussion in Sec. III of this paper.

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Superconducting Transition Temperatures and Lattice Parameters of Simple-Cubic Metastable Te-Au Solutions Containing Fe and Mn[†]

L. R. Newkirk and C. C. Tsuei

W. M. Keck Laboratory of Engineering Materials, California Institute of Technology, Pasadena, California 91109

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The superconducting transition temperatures and lattice spacings of simple-cubic Te-Au-Fe and Te-Au-Mn alloys, prepared by rapid quenching from the liquid state, have been measured and correlated with anomalies in the Te-Au system and the band structure proposed to explain those anomalies. The unusual behavior of these properties in the ternary alloys containing Fe and Mn has been interpreted in terms of a Fermi-surface-Brillouin-zone interaction based on information obtained from studying binary Te-Au alloys. The results of this study lend additional support to the electronic band structure proposed for simple-cubic Te-Au alloys, and in addition show a very distinct band-structure effect on the superconducting transition temperatures of the Te-Au-Fe alloys.

I. INTRODUCTION

A recent investigation¹ has shown that the anomalies in the variation of lattice parameter, thermoelectric power, and superconducting transition temperature with concentration in liquid-quenched sim-

ple-cubic Te-Au alloys varying in composition from 60 to 85 at. % Te can be qualitatively explained in terms of a Fermi-surface-Brillouin-zone interaction. Subsequently it was found that about 2 at. % Mn and about 7.5 at. % Fe could be retained in solid solution in quenched Te-Au alloys. The effect of

these magnetic impurity atoms on the lattice parameter and on the superconducting transition temperature is the object of the present investigation.

II. EXPERIMENTAL PROCEDURE

The alloys were prepared and quenched from the liquid state following the technique described in Ref. 1. Lattice parameter measurements were obtained from Debye-Scherrer diffraction patterns ($\text{CuK}\alpha$ radiation) and the Nelson-Riley extrapolation function. Iron was introduced in several binary Te-Au alloys containing from 20 to 40 at. % Au, while manganese was introduced only in the base alloy $\text{Te}_{70}\text{Au}_{30}$. Preliminary experiments were performed in order to determine the maximum concentration of either Fe or Mn which would be retained in the simple-cubic phase after quenching, as evidenced by the absence of diffraction peaks other than those of the simple-cubic structure. Within the single-phase region, the lattice parameters (as shown later) varied monotonically with Fe or Mn concentration. A list of the alloy compositions investigated is given in Table I. As shown in this table, the maximum Fe concentration that could be added to the binary compositions Te-Au was about 7.5 at. % except for the $\text{Te}_{80}\text{Au}_{20}$ base alloy for which only 2.5% could be introduced. The maximum concentration of Mn was only 2 at. % in the $\text{Te}_{70}\text{Au}_{30}$ base alloy. The superconducting transition temperature was measured by a standard ac bridge technique, using a frequency of 1 kHz. Temperature was measured with a Honeywell germanium resistance thermometer calibrated against the 1958 He^4 vapor pressure scale of temperature.²

In analyzing the effect on the superconducting transition temperature, of either Fe or Mn in solid solution in the simple-cubic structure, it was important to know whether or not these atoms had a magnetic moment. A direct measurement of these moments at low temperature with the available magnetometer would have required at least 200 mg of each alloy. Since the average weight of a quenched foil is about 1 mg, and each foil must be subjected to a very accurate x-ray diffraction experiment in order to establish that it contains only the simple-cubic phase, such measurements were judged impractical.

In the absence of magnetometer data some evi-

TABLE I. Composition of alloys.

Alloy system	x(at. %)
$(\text{Te}_{80}\text{Au}_{20})_{100-x}\text{Fe}_x$	2.5
$(\text{Te}_{75}\text{Au}_{25})_{100-x}\text{Fe}_x$	0.5, 1.0, 1.5, 2.0, 2.5, 3.75, 6.25, 7.5
$(\text{Te}_{70}\text{Au}_{30})_{100-x}\text{Fe}_x$	0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 5.0, 7.0
$(\text{Te}_{65}\text{Au}_{35})_{100-x}\text{Fe}_x$	2.5, 5.0, 7.5
$(\text{Te}_{60}\text{Au}_{40})_{100-x}\text{Fe}_x$	2.5, 5.0, 7.5
$(\text{Te}_{70}\text{Au}_{30})_{100-x}\text{Mn}_x$	0.5, 1.0, 1.5, 2.0

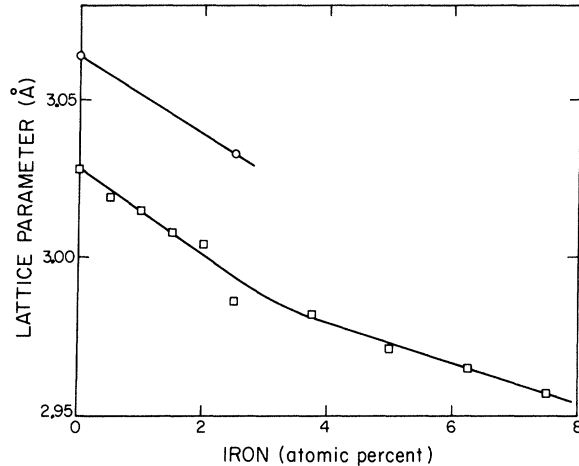


FIG. 1. Lattice parameter of the simple-cubic alloys $(\text{Te}_{80}\text{Au}_{20})_{100-x}\text{Fe}_x$ (○), and $(\text{Te}_{75}\text{Au}_{25})_{100-x}\text{Fe}_x$ (□).

dence to support the conclusions regarding localized moments to be presented later may be obtained by measuring the variation of magnetoresistance with magnetic field. Beal-Monod and Weinert³ have shown in relation to the Kondo effect that a negative magnetoresistance is associated with the existence of a localized magnetic moment. For these measurements, a constant current was obtained from a Princeton Applied Research current source and the potential was measured by a Leeds and Northrup guarded potentiometer. The magnetic field was measured by a Varian proton resonance meter.

III. RESULTS

The results of lattice parameter measurements are shown in Figs. 1 and 2 for the alloys containing Fe and in Fig. 3 for those containing Mn. In all cases the lattice parameter of the simple-cubic alloys decreases smoothly with increasing concentrations of either Fe or Mn. The alloys based on $\text{Te}_{75}\text{Au}_{25}$ are of particular interest since the lattice spacings seem to fall on two segments of straight lines as shown in Fig. 1.

The superconducting transition temperatures for alloys containing Fe were measured only for alloys based on $\text{Te}_{75}\text{Au}_{25}$, $\text{Te}_{70}\text{Au}_{30}$, and $\text{Te}_{65}\text{Au}_{35}$ and are presented in Figs. 4-6. No graphical results are presented for the alloys containing Mn, since it was found that the transition temperature was below 1.3 °K for a Mn concentration of only 0.5 at. %. The magnetoresistance of the alloys based on $\text{Te}_{70}\text{Au}_{30}$ and containing 1.5 at. % of either Fe or Mn are shown in Fig. 7. In spite of the scatter in the results, especially below a field of 4 kG, it is obvious that the magnetoresistivity is positive for the alloy containing Fe, and negative for that containing Mn.

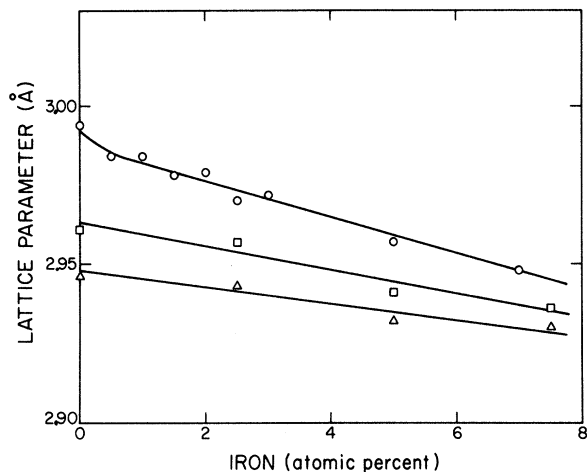


FIG. 2. Lattice parameter of the simple-cubic alloys $(Te_{70}Au_{30})_{100-x}Fe_x$ (○), $(Te_{65}Au_{35})_{100-x}Fe_x$ (□), and $(Te_{60}Au_{40})_{100-x}Fe_x$ (△).

IV. DISCUSSION

An important consideration in analysing the functional dependence of the superconducting transition temperature on the concentration of magnetic impurity is the presence or absence of a localized magnetic moment associated with the impurity atom. The basic qualitative argument concerning the formation of a localized moment is due primarily to Friedel.⁴ He argues that when an element of the first transition series is added to a solvent, the $3d$ energy levels of the impurity resonate with the conduction states of the solvent. This causes a broadening of the $3d$ band of the impurity and a relative shift between the $3d \uparrow$ band and the $3d \downarrow$ band.

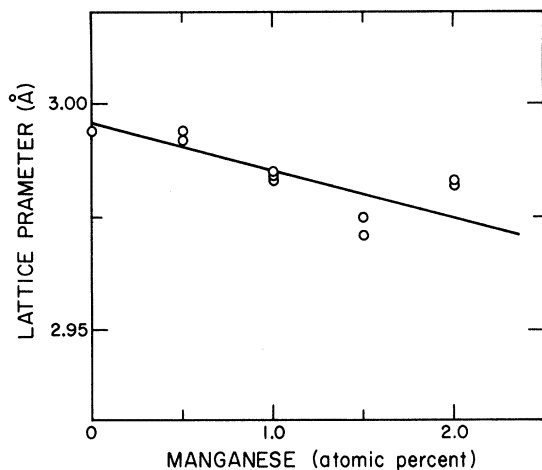


FIG. 3. Lattice parameter of the simple-cubic alloys $(Te_{70}Au_{30})_{100-x}Mn_x$.

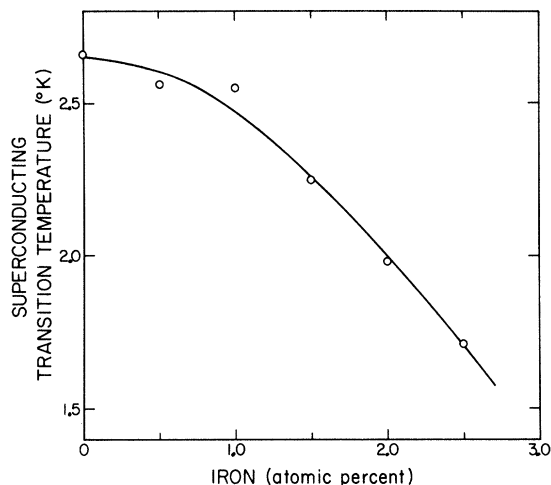


FIG. 4. Superconducting transition temperature of the simple-cubic alloys $(Te_{75}Au_{25})_{100-x}Fe_x$. Each point represents the average of at least four specimens.

If the Fermi level of the host lies entirely above or below the impurity levels, then both the $3d \uparrow$ and the $3d \downarrow$ bands will be completely full or empty, and the impurity will exhibit no localized moment. If, however, the Fermi level falls in the region of the impurity levels, then due to the shift between bands of opposite spin, there will be unequal numbers of $3d \uparrow$ and $3d \downarrow$ electrons, and the impurity will exhibit a localized magnetic moment.

The Fermi energy levels for two Te-Au binary alloys were calculated by assuming the free-electron model. The results were 8.4 eV for $Te_{60}Au_{40}$ and 8.9 eV for $Te_{85}Au_{15}$. According to Ref. 5, the top of the $3d$ band for Fe seems to be about 7 eV,

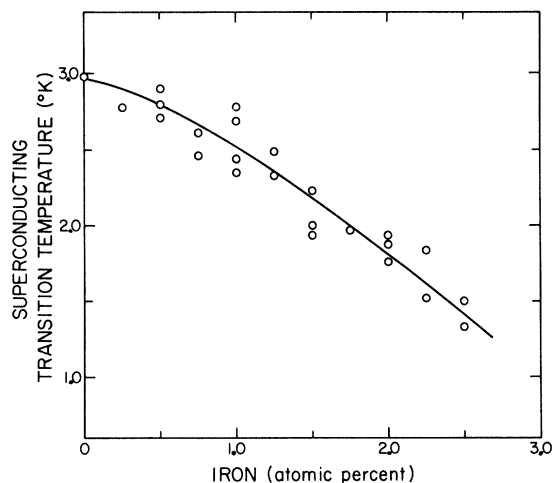


FIG. 5. Superconducting transition temperature of the simple-cubic alloys $(Te_{70}Au_{30})_{100-x}Fe_x$.

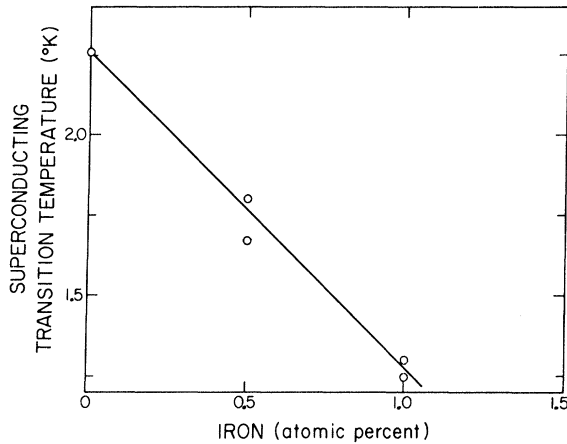


FIG. 6. Superconducting transition temperature of the simple-cubic alloys $(\text{Te}_{65}\text{Au}_{35})_{100-x}\text{Fe}_x$.

and between 10 and 12 eV for Mn. On the basis of this information and Friedel's model, Fe dissolved in the two above mentioned Te-Au alloys would not be expected to have a localized magnetic moment, but Mn would. This is consistent with the results of magnetoresistance measurements shown in Fig. 7.

It is known that the superconducting transition temperature of solid-solution alloys is generally sensitive to the concentration of a solute element having a localized moment, and according to Ref. 6 the variation of superconducting transition temperatures with solute concentration can in many cases be a very sensitive test for magnetic states. The slopes of the T_c vs concentration curves for several binary Te-Au alloys to which Fe was added

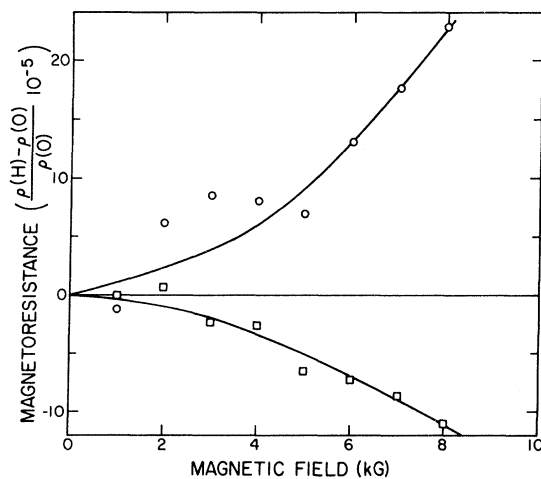


FIG. 7. Transverse magnetoresistance of the simple-cubic alloys $(\text{Te}_{70}\text{Au}_{30})_{98.5}\text{Fe}_{1.5}$ (O), and $(\text{Te}_{70}\text{Au}_{30})_{98.5}\text{Mn}_{1.5}$ (□).

range between 0.4 and 1.0 °K/at. % Fe, while that for Mn added to the alloy $\text{Te}_{70}\text{Au}_{30}$ is greater than 3.5 °K/at. % Mn. A summary of available data for superconductors containing elements of the first transition series is shown in Table II. This information suggests that an increase of an order of magnitude or more in the rate of depression of the critical temperature with increasing solute concentration may be expected if the solute has a magnetic moment. The results of the present investigation are consistent with this general observation.

In many problems concerning alloys, electron concentrations are often more meaningful than atomic concentrations. In binary Te-Au alloys, valences of 4 for Te and 1 for Au have been suggested in Ref. 1. The valence of the Fe atom entering in solid solution in the simple-cubic structure of the binary Te-Au alloys is open to question. Since Fe in these alloys does not have a localized moment, it may be concluded that the conduction electrons from Te and Au entirely fill the localized d band of the Fe atoms, and if so, four conduction electrons would be removed from the Fermi sea for each Fe atom in solid solution. The effect of the 4s electrons of Fe is more difficult to analyze. For certain alloys involving Fe and Co, Wilson⁷ concludes that the 4s electrons join the conduction band, resulting in a valence of -2 for Fe. On the other hand, APW calculations for bcc Fe⁸ show that the 4s band is well below the 3d band and does not even extend close to the Fermi surface. Unfortunately there is no evidence of what happens to the 4s band when Fe is dissolved in Te-Au, so the decision to assign a valence of -4 to Fe was based on the resulting con-

TABLE II. Superconducting transition temperatures of solid solutions of Zn, Al, and In with transition metals.

Alloy system	dT_c/dc (°K/at. % x)	Magnetic moment
$\text{Zn}_{100-x}\text{Cr}_x$	-180 a	Yes (Ref. 14)
$\text{Zn}_{100-x}\text{Mn}_x$	-310 a	Yes b, c
	-170 (Ref. 11)	
$\text{Zn}_{100-x}\text{Fe}_x$	-13 a	Unknown
$\text{Zn}_{100-x}\text{Co}_x$	-6.4 a	Unknown
$\text{Zn}_{100-x}\text{Ni}_x$	-2.9 a	Unknown
$\text{Al}_{100-x}\text{Cr}_x$	-2.7 (Ref. 11)	No (Ref. 14)
$\text{Al}_{100-x}\text{Mn}_x$	-7.3 (Ref. 11)	No d, e
$\text{Al}_{100-x}\text{Fe}_x$	-2.4 (Ref. 11)	No d, e
$\text{In}_{100-x}\text{Mn}_x$	-6 (Ref. 12)	No (Ref. 12)

^aD. L. Martin, Proc. Phys. Soc. (London) **78**, 1489 (1961).

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sistency in the interpretation of the experimental findings.

Superconducting Transition Temperatures

The superconducting transition temperature of Te-Au-Fe alloys varies in an unusual manner as the concentration of Fe is increased (Figs. 4 and 5). The dependence of T_c on Fe concentration shows a negative curvature in the region of low Fe concentrations which is seemingly inconsistent with the more widely accepted theories.⁹⁻¹¹ As pointed out above, however, Fe added to Te-Au alloys behaves as though it had a valence of approximately -4. This implies that the addition of small amounts of Fe will cause the Fermi surface to move relatively large distances in momentum space. Therefore, before the impurity effect of Fe on the transition temperature can be compared with theoretical predictions, it is necessary to correct for the effect of the changing position of the Fermi surface. It is possible to separate these two effects on the basis of the information obtained from the superconducting transition in Te-Au alloys and the band-structure model presented in Ref. 1. In order to do this it must be assumed that (i) compositional dependence of the transition temperature of pure Te-Au is due entirely to a changing Fermi surface within a rigid-band model and (ii) the addition of 2-3 at. % Fe does not significantly alter the band structure. Based on these assumptions, the following correction procedure can be applied. The concentration of electrons per atom is computed for the Te-Au-Fe alloy being corrected. The composition of the Te-Au alloy which has the same electronic concentration is then determined, and the superconducting

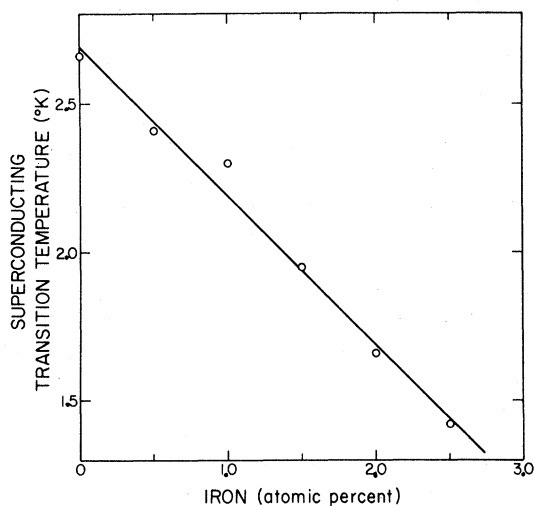


FIG. 8. Superconducting transition temperature of the simple-cubic alloys $(\text{Te}_{75}\text{Au}_{25})_{100-x}\text{Fe}_x$ after correction described in Eq. (1).

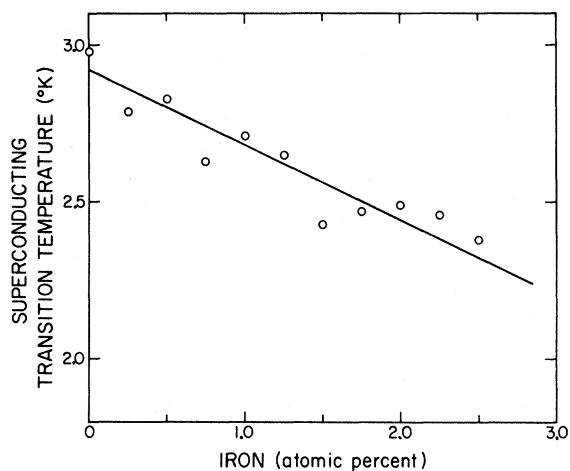


FIG. 9. Superconducting transition temperature of the simple-cubic alloys $(\text{Te}_{70}\text{Au}_{30})_{100-x}\text{Fe}_x$ after correction described in Eq. (1).

transition temperature of that alloy is obtained from Fig. 2 of Ref. 1. This temperature T_{c1} is the transition temperature one would expect if the addition of Fe had no effect other than to change the location of the Fermi surface. The actual observed transition temperature of the Te-Au-Fe alloys T_{c2} is in all cases lower than T_{c1} , implying that either disordering or magnetic properties of the Fe lowered the superconducting transition temperature by an amount $\Delta T = T_{c1} - T_{c2}$. The corrected transition temperatures shown in Figs. 8-10 were obtained by the formula

$$T_c(x) = T_{c0} - \Delta T(x), \quad (1)$$

where T_{c0} is the transition temperature of the binary Te-Au alloy to which Fe was added, and x is

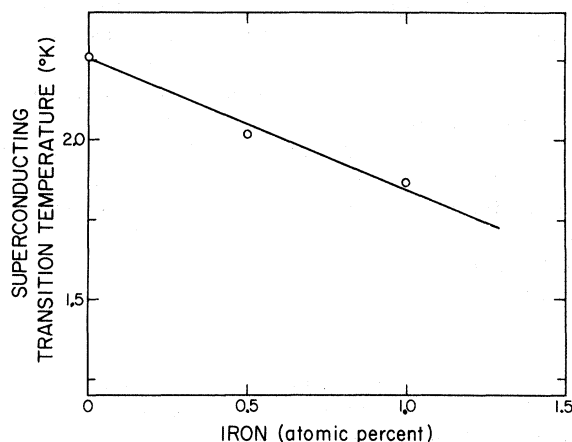


FIG. 10. Superconducting transition temperature of the simple-cubic alloys $(\text{Te}_{65}\text{Au}_{35})_{100-x}\text{Fe}_x$ after correction described in Eq. (1).

the Fe concentration. The correction is obtained for each alloy as follows: The alloy $(\text{Te}_{75}\text{Au}_{25})_{98.5}\text{Fe}_{1.5}$, for example, becomes superconducting at 2.25 °K (Fig. 4). The electron per atom ratio computed on the basis of $\text{Au} = +1$, $\text{Te} = +4$, and $\text{Fe} = -4$ is 3.14 electrons per atom, which in the binary Te-Au system corresponds to the alloy $\text{Te}_{71.3}\text{Au}_{28.7}$. The transition temperature of this alloy is 2.95 °K (Fig. 2 of Ref. 1), hence, the impurity effect of Fe is to lower the transition temperature by an amount $\Delta T = 2.95 - 2.25 = 0.70$ °K. The transition temperature of the $\text{Te}_{75}\text{Au}_{25}$ base alloy is 2.66 °K (Fig. 2 of Ref. 1); therefore, the corrected transition temperature for this alloy is given by

$$T_c = T_{c0} - \Delta T = 2.66 - 0.70 = 1.96 \text{ °K.}$$

A theory of the effect of nonmagnetic impurities on the superconducting transition temperature of elements has been proposed by Markowitz and Kadannoff¹⁰ and will be used to interpret the corrected transition temperatures presented here. The main conclusion of this theory is the prediction that the functional dependence of the transition temperature on impurity concentration should be of the following form:

$$T_c = T_{c0} - K^i X - \langle a^2 \rangle T_c I_c(X), \quad (2)$$

where K^i is a constant depending on the host, X is a variable proportional to concentration, $\langle a^2 \rangle$ is a factor which depends on the anisotropy of the electron-phonon pairing potential, and $I_c(X)$ is a fairly complicated integral which will not be required for this discussion. According to these authors, the term $\langle a^2 \rangle T_c I_c(X)$ is due to the reduction in anisotropy resulting from a reduction in the mean collision time, and the linear term $K^i X$ is due to a change in the gross properties of the system. The above formula was derived for the addition of an impurity to an elemental superconductor, in which case small amounts of impurity will greatly reduce the mean collision time and smooth out the anisotropy of the pairing potential. In the present case the host material is already a completely disordered solid solution of Te and Au and it seems unlikely that the addition of a few Fe atoms will have any substantial effect on the mean collision time. Hence, the last term in Eq. (2) may be disregarded and the functional dependence of the transition temperature reduces to the form

$$T_c = T_{c0} - K^i \lambda x, \quad (3)$$

where λ is the proportionality constant between X and the impurity concentration x . As shown in Figs. 8–10 the corrected transition temperatures do indeed behave in the manner described by this equation. In evaluating this explanation of the behavior of the superconducting transition tempera-

ture, it should be realized that (i) the theory was developed using an elemental and not a solid-solution host, with the result that some of the parameters, such as mean collision time, may not vary in the manner assumed, and (ii) the theory was intended to apply to cases where the transition temperature was altered by a few percent, and not by the larger amount occurring in the alloys studied.

The shapes of the T_c -vs- x curves for several alloy systems where the host material is an elemental superconductor and the solute is a metal from the first transition series (without a detectable localized moment) seem to support this explanation. The systems Al-Cr and Al-Mn¹² appear to show a definite curvature of the type predicted by Eq. (2). The work by Martin¹³ on the system In-Mn also suggests this same mechanism although the amount of Mn in solid solution may be open to question as a result of the technique of alloy preparation. The work of Opitz¹⁴ on the system In-Fe shows a linear relationship between T_c and Fe concentration very similar to that observed in this study. His samples were prepared by vapor deposition which probably introduces many imperfections into the crystal structure. This is likely to have the same effect as disordering in an alloy, since it will reduce the mean free path and probably smooth out the anisotropy in the energy gap. If this is the case, a linear relationship such as that observed is quite consistent with this interpretation.

Lattice Parameters

The variation of the lattice parameters of the binary Te-Au simple-cubic alloys with concentration can be approximated by two straight lines with a change of slope around 68 at. % Te. This anomaly was explained in Ref. 1, as due to the critical concentration at which the Fermi surface makes contact with the second Brillouin zone. Assuming that the valences previously discussed for Te and Au are correct (4 and 1, respectively), the change in slope in the Te-Au alloys occurs at an electron concentration of about 3.04 electrons per atom. A purely geometric construction based on the free-electron model indicates contact between the Fermi surface and the Brillouin zone at 2.96 electrons per atom, which is in satisfactory agreement with the experimental findings. The concentration of 68 at. % Te however, is very close to that at which the compound Te_2Au is found in the equilibrium alloys, and the existence of this compound may also be a reason for the anomaly in the lattice parameters of the simple-cubic phase. The results of the present investigation give additional evidence for the Fermi-surface-Brillouin-zone contact interpretation. When Fe is present in solid solution in the Te-Au binary alloys, changes in slopes are also observed as shown in Figs. 1 and 2. These data were replotted in terms

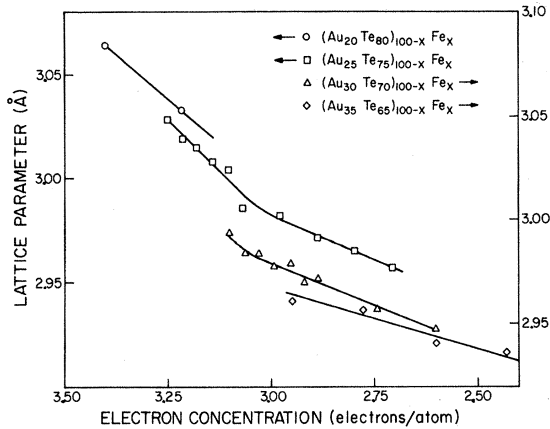


FIG. 11. Lattice parameter of the simple-cubic Te-Au-Fe alloys as a function of electron per atom ratio.

of electron concentrations and are shown in Fig. 11. It is apparent that the change in slope observed oc-

curs at an electron concentration of approximately 3 electrons per atom. It may be concluded from this that the anomaly in lattice parameters is due to the effect of conduction electrons and is not influenced by the existence of the Te_2Au compound in equilibrium alloys. This conclusion demonstrates that a lattice parameter anomaly of the type occurring in many hexagonal alloys^{15,16} can also occur in cubic systems. In addition it seems that this effect depends only on the concentration of conduction electrons and not on alloy composition. Considering the independent evidence showing that the Fermi surface contacts a Brillouin-zone boundary where the lattice parameter anomaly occurs, the conclusion that the lattice spacings in a cubic system may be influenced by this contact is clearly suggested.

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