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THE EFFECT OF IMPURITIES ON THE SUPERCONDUCTING TRANSITION TEMPERATURE IN THALLIUM

T. A. IGNAT'EVA, V. I. MAKAROV, and N. S. TERESHINA

Physico-technical Institute, Academy of Sciences, Ukrainina S.S.R.

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Binary and ternary solid solutions of thallium containing bismuth, antimony, indium, and mercury are studied. It is found that the quantity dT_c/dP as a function of the impurity concentration has a maximum. Such a dependence indicates that the topology of the Fermi surface changes under the influence of the impurity.

IT has been established in a number of papers that the pressure dependence of the superconducting transition temperature of thallium is extremely sensitive to the valence of the impurity atom.[1-4] The obtained experimental data were explained by a change in the topology of the Fermi surface of thallium under the influence of the impurity and pressure.[5,6] In this paper these ideas have been further developed on the basis of new experimental material: the effect of one and two impurities (binary and ternary solid solutions) on the superconducting transition T_c of thallium, its derivative with respect to pressure dT_c/dP, and the volume changes of the unit cell of thallium under the influence of the impurity.

PREPARATION OF SAMPLES AND THE METHOD OF MEASUREMENT

We have investigated the effect of mercury impurities on the value of T_c of solid solutions of thallium with antimony and indium impurities (ternary solid solutions), as well as the effect of bismuth and antimony impurities on the value of T_c of pure thallium. The following ternary solutions were prepared for investigation.

Tl – Sb(0.042 at.%) – Hg, Tl – Sb(0.084 at.%) – Hg,
Tl – In(1at.%) – Hg, Tl – In(2at.%) – Hg.

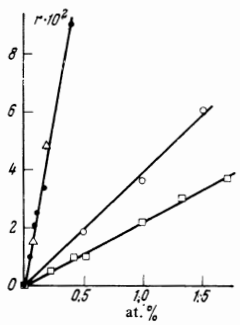

FIG. 1. The dependence of the residual resistance of thallium on the impurity concentration: ●–Bi, Δ–Sb, ○–Hg, and □–In. [4]

The concentration of the mercury impurity varied from 0 to 1.2 at. percent. In order to prepare good, uniform ternary solutions, we first prepared binary solutions with given content of antimony or indium impurities and then ternary solutions with the maximum content of mercury impurity (1.2 at. percent). Ternary solutions with lower mercury content were prepared by dilution of the maximum solution by the initial binary solution.

The maximum mercury content was monitored by the relative residual resistance

$$r = R(4.2^\circ\,\mathrm{K}) / R(300^\circ\,\mathrm{K}),$$

which consists of the residual resistance of each im-

purity. For each impurity in the investigated region of concentrations the value of r depends linearly on the concentration (Fig. 1).

The resistance was measured by the potentiometer method. The ice method[7] was used to produce the pressure.

THE RESULTS OF THE MEASUREMENTS

1. The effect of impurities on the value of T_c of thallium. From investigations of the effect of bismuth, antimony, and indium on T_c of thallium (Fig. 2) it is seen that starting with the lowest concentrations indium and bismuth increase the value of T_c of thallium in the entire range of investigated concentrations. Under the action of the antimony impurity the value of T_c of thallium first decreases inappreciably (to $\sim 7 \times 10^{-3}$ deg) and then increases, as in the case of indium and bismuth impurities, with further increase in the impurity concentration.

In investigations of the ternary solid solutions Tl-Sb-Hg and Tl-In-Hg it turned out that the nature of their dependence of ΔT_c on the mercury impurity concentration (Fig. 3, curves 2–5) does not differ from that of pure thallium with mercury impurity (curve 1). Independently of the initial impurity content of antimony or indium, mercury decreases the value of T_c of thallium in the entire range of investigated concentrations. The behavior of $T_c(\Delta T_c)$ is similar in all the solid solutions.

2. The effect of impurities on the value of T_c of thallium under pressure. We have investigated in detail the joint effect of impurities and pressure on the superconducting transition temperature for ternary solutions (Fig. 4). The initial points of the curves on Fig. 4 correspond to the change in the superconducting transition temperature under a pressure of 1730 kg/cm^2 for the binary solutions Tl-Sb (0.042 at. percent), Tl-Sb (0.084 at. percent), and Tl-In (1 at. percent). Under the influence of mercury impurities the value of ΔT_c^P of these binary solid solutions changes from zero or from a negative value, passes through a positive maximum, and becomes negative on further increasing the mercury concentration.

Figure 5 presents the pressure dependence of ΔT_c for the binary solution Tl-In (1 at. percent) with two mercury concentrations in the pressure range 0–1730 kg/cm^2. It is seen that the dependence $\Delta T_c(P)$ for a ternary solution is not linear.

3. X-ray structure investigations of the solid solutions of thallium. All the investigated solid solutions, as well as pure thallium, have a hexagonal structure with a c/a axial ratio close to 1.6. The solid solutions were investigated using the back-reflection method and copper radiation. Their parameters were determined from the $K\alpha_1$ and $K\alpha_2$ doublets of the (125) and (206) lines. Pure thallium with a = 3.4565 Å and c = 5.5249 Å served as the standard. The accuracy of the determination of a and c amounted to $\pm 7 \times 10^{-4}$ Å.

The results are shown in Fig. 6. As is seen from the figure, for the binary solutions Tl-In and Tl-Hg the a parameter decreases appreciably, almost equally for both solutions, whereas the c parameter remains almost unchanged. The change of the parameters in the ternary solution Tl-In (1 at. %)-Hg is of the same nature. In all cases there is an increase in the c/a ratio.

In the case of thallium with an antimony concentration the dependence of a and c on the impurity concentration is different. With increasing antimony content a decrease is observed in the values of a and c to a minimum value (up to $r = 4 \times 10^{-2}$ or 0.13 at. percent Sb); with further increasing impurity concentration one observes a smooth increase of the parameters. The c/a ratio decreases.

DISCUSSION OF THE RESULTS

Before we turn to a discussion of the results, we note that the nonlinear dependences of T_c on the impurity concentration and pressure can be considered as the sum of two components: a linear and a nonlinear component.[5,6,8,9] The linear dependence is in both instances connected with linear corrections to the density of electron states $\nu(\epsilon_F)$ (ϵ_F is the Fermi energy) and to the Debye temperature ω_D. As will be seen below, the cited experimental data show that the nonlinear dependences of T_c and of dT_c/dP on the impurity con-

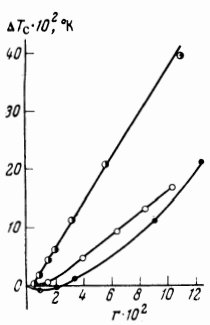

FIG 2.

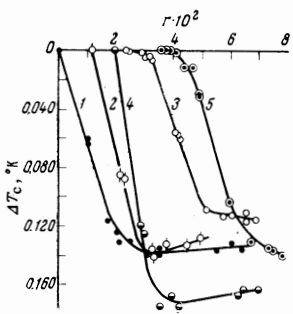

FIG 3.

FIG 2. The shift of T_c of thallium under the influence of impurities: ◑–In, [4] ○–Bi, and ●–Sb.

FIG. 3. The shift of T_c of solid solutions of thallium with indium and antimony under the influence of mercury impurities: curve 1–Tl + Hg, 2–Tl + 0.042 at. % Sb + Hg, 3–Tl + 0.084 at. % Sb + Hg, 4–Tl + 1 at. % In + Hg, 5–Tl + 2 at. % In + Hg.

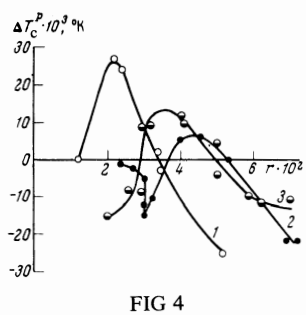

FIG 4

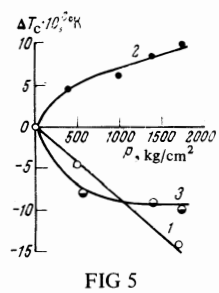

FIG 5

FIG. 4. The dependence of $\Delta T_c P$ of ternary solid solutions of thallium on r (Hg concentration) at a pressure of 1730 kg/cm^2. Curve 1–Tl + 0.042 at. % Sb + Hg, 2–Tl + 0.084 at. % Sb + Hg, 3–Tl + 1 at. % In + Hg.

FIG. 5. The $\Delta T_c(P)$ dependence in a solid solution of Tl + 1 at. % In at various concentrations of Hg: curve 1–Tl + 1 at. % In, 2–Tl + 1 at. % In + 0.5 at. % Hg, 3–Tl + 1 at. % In + 1 at. % Hg.

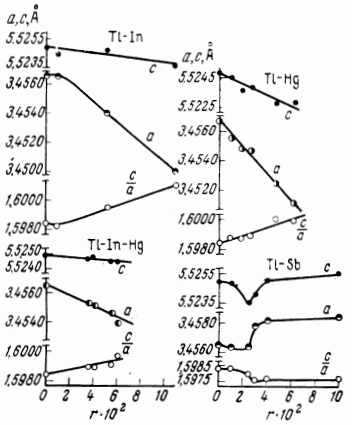

FIG. 6. The dependence of the lattice parameters of thallium on r (the amount of impurity) for binary and ternary solid solutions.

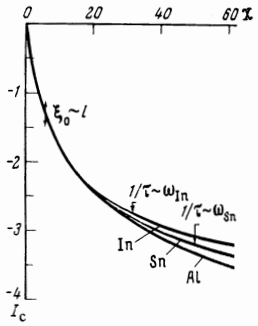

FIG. 7

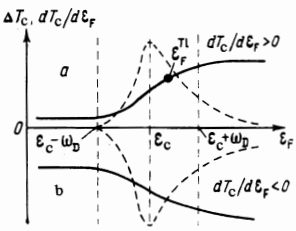

FIG. 8

FIG. 7. The dependence of I_c on χ. The arrows indicate the values of χ for which $1/\tau \sim \omega_D$ of the corresponding metals.

FIG. 8. The dependence of ΔT_c (solid lines) and of $dT_c/d\epsilon_F$ (dashed line) on ϵ_F in the simplest topological cases: a—production of one of the portions of the Fermi surface, b—disappearance of one of the portions of the Fermi surface. ϵ_c is the critical energy at which the change in the topology of the Fermi surface occurs, and ϵ_F^{Tl} is the Fermi energy of thallium.

centration for thallium can basically be explained by one and the same mechanism—a change in the typology of the Fermi surface under the influence of the impurity.

Let us consider the two existing points of view of the nature of the nonlinear mechanisms of the change of T_c as a function of the pressure and the impurity concentration [5,6,9]. One of them is a decrease of the anisotropy of the energy gap due to the impurity.[9] In the isotropic model T_c does not depend on the impurity concentration.[10] However, if the electron-electron interaction is anisotropic and of the form

$$I(\mathbf{p}, \mathbf{p}') = [1 + a(\hat{\Omega})]I_0[1 + a(\hat{\Omega}')],$$

where $a(\hat{\Omega})$ is the anisotropic part of the interaction, then the temperature of the superconducting transition will change and the equation from which T_c is determined takes on the following form [9]:

$$1 = I_0 \nu(\varepsilon_F) \int_0^{\omega_D} \text{th}\frac{\omega}{2T_c}\left[1 + \frac{\langle a^2\rangle}{1+(2\omega\tau_a)^{-2}}\right]\frac{d\omega}{\omega}, \qquad (1)$$

where τ_a is the smearing time of the anisotropic part of the electron-electron interaction.

Assuming $\langle a^2 \rangle \ll 1$, one can write the change of the superconducting transition temperature for different τ_a as follows:

$$\Delta T_c(\chi) = T_c\langle a^2\rangle I_c(\chi), \quad \chi = (T_c\tau_a)^{-1}. \qquad (2)$$

In the case of an anisotropic scattering center

$$\chi = \frac{\tau}{\tau_a} = \frac{\xi_0}{0.18b}r,$$

where ξ_0 is the coherence parameter, $b = rl$, $l \propto \tau$ is the mean free path of the electron on the Fermi surface, and τ is the mean free time. The value of χ is proportional to the impurity concentration.[9] It is significant that the shift ΔT_c depends on the total amount of the impurity and does not depend on its valence.[9] Figure 7 presents the dependence of I_c on χ.[9]

It can be shown that the quantity dT_c/dP as a function of χ, and consequently also of the impurity concentration, has neither a maximum nor a minimum. Differentiating Eq. (1), we obtain

$$\frac{1}{T_c}\frac{dT_c}{dP} = \left(\frac{1}{T_c}\frac{dT_c}{dP}\right)_0 + I_c(\chi)\frac{d\langle a^2\rangle}{dP}, \qquad (3)$$

where the first term on the right corresponds to an isotropic superconductor and does not depend on the parameter χ, and the dependence of the second term on χ is seen from Fig. 7.

Another possibility of explaining the nonlinear change of T_c as a function of the impurity concentration and pressure are the topological changes in the electron spectrum of the metal.[5,6] In that case the change in the superconducting transition temperature is

$$\Delta T_c = T_c F(\varepsilon_F),$$

where the form of the function $F(\epsilon_F)$ depends on the nature of the topological transition. Unlike in the case of the first mechanism, when T_c only decreases, in this case both a decrease and an increase in T_c is possible. For instance, Fig. 8 shows the form of $T_c(\epsilon_F)$ for two specific topological instances—the production of a small portion of the Fermi surface (a) and its disappearance (b). As has been shown in [5], dT_c/dP as a function of ϵ_F (Fig. 8), and consequently also of the impurity concentration, can have either a maximum or a minimum.

The cited experimental data on Figs. 2–6 make it possible to clarify which of the two nonlinear mechanisms is the determining one in the dependence of T_c and dT_c/dP on the impurity concentration. The decrease in the value of dT_c/dP in the solutions Tl-In, Tl-Bi, and Tl-Sb (Fig. 4) could be explained by a decrease in the anisotropy of the gap under the influence of the impurity [expression (3)].

If we plot the dependence of $\Delta T_c/r$ on ln r for the investigated solutions under pressure and without pressure then one can draw conclusions regarding the change in the anisotropy of the gap $\langle a^2 \rangle$ under pressure from the change of the slope (Fig. 9). Assuming $\tau/\tau_a = 1$ for the indium impurity, one can determine $\langle a^2 \rangle$ which turned out from our data to be 0.058. If we disregard the data on thallium with the antimony impurity, under the influence of which the lattice parameter changes in a complicated manner (Fig. 6), then the change of $\langle a^2 \rangle$ under the influence of a pressure of 2000 kg/cm^2 amounts to no more than 10 ± 10 percent. The possible increase of the gap anisotropy at that pressure amounts then according to our calculations to 6×10^{-3}.

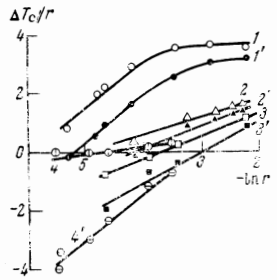

FIG. 9

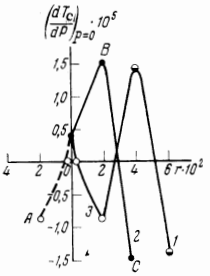

FIG. 10

FIG. 9. The dependence of $\Delta T_c/r$ on ln r in: 1, 1′–Tl + In, 2,2′–Tl + Bi, 3, 3′–Tl + Sb. Curves 1–3 at P = 0, curves 1′–3′ at P = 1730 kg/cm². Curves 4 and 4′–data from the work of Gey [11] at P = 0 and P = 2000 kg/cm².

FIG. 10. The dependence of $(dT_c/dp)_{p=0}$ on r for solid solutions of thallium: 1–Tl + 1at.% In with various mercury contents, 2–Tl + Hg [1], 3–Tl + In. Dashed line–reflection of curve 3.

Let us attempt to explain the entire aggregate of experimental data, assuming that the nonlinear component of the pressure effect is due to a change in the gap anisotropy under pressure. We will assume, as before, that in thallium there appear under pressure two components[1]–a linear component [ΔT_c^l with $dT_c/dP = -1.4 \times 10^{-5}$ deg/(kg/cm²)[1]] and a nonlinear one (ΔT_c^n). Then, calculating

$$\Delta T_c^l\,(P = 2000\ \text{kg/cm}^2) = -28\cdot 10^{-3^\circ}\,\text{K}$$

$$\Delta T_c^n(P = 2000\ \text{kg/cm}^2) = \frac{T_c}{I_0 \nu(\varepsilon_F)}\Delta\langle a^2\rangle = +45\cdot 10^{-3^\circ}\,\text{K},$$

we find for pure thallium

$$\Delta T_c = \Delta T_c^n + \Delta T_c^n = +17\cdot 10^{-3^\circ}\,\text{K},$$

which is in fairly good agreement with the experimental value of $15 \times 10^{-3^\circ}$K obtained previously.[2]

It would appear that we have thereby explained the nonlinear change of $\Delta T_c(P)$ in thallium. However this point of view is contradicted by the presence of a maximum in the dependence of dT_c/dP on the impurity concentration for the solutions Tl-Hg, Tl-In-Hg, and Tl-Sb-Hg (Fig. 4). Instead of the decrease in dT_c/dP expected according to the theory of Markowitz and Kadanoff[9] [expression (3)], one observes experimentally an increase of dT_c/dP (Fig. 4). On increasing the impurity concentration the derivative dT_c/dP goes through a maximum (Fig. 10; in plotting these dependences we used the data of Fig. 5).

The qualitative agreement of the experimental data of the previously published work of Gey with the theory[9] appears to us to be accidental. In that work he investigated the dependence of ΔT_c for pure thallium on the residual resistance r produced by plastic deformation at various pressures. Figure 9 shows the results of this work treated in accordance with the theory of Markowitz and Kadanoff.[9] It is seen that according to the data of Gey's work the anisotropy of the gap $\langle a^2\rangle$ (the slope) changes by a factor of four at a pressure of 2000 kg/cm² compared with P = 0 (curves 4 and 4′), which is in disagreement with our data [$\Delta\langle a^2\rangle/\langle a^2\rangle \sim 10$ percent]. It appears to us that such a discrepancy in the results is connected with the following circumstances. First, the effect of the residual resistance produced in plastic deformation upon T_c is not equivalent to the effect of the residual resistance produced by impurities.[12] Secondly, plastic deformation results in a complex system of stresses which can change strongly when an external pressure is applied and this will manifest itself in the dependence of ΔT_c^P on r. We note also that a large arithmetical error has been found[13] in Gey's calculations of the proportionality coefficient between χ and r which is important for the comparison with theory. Thus the theory of Markowitz and Kadanoff[9] does not explain the entire aggregate of the existing experimental data.

It should be noted that the presence of a maximum in the dependences of dT_c/dP on the concentration in thallium solutions with impurities is not related with any peculiarities in the volume changes of the unit cell of thallium under the influence of the impurity. Thus, in the binary solutions Tl-In and Tl-Hg these changes are identical in magnitude and sign. However, a sharp discontinuity in the course of the change of the parameters as a function of the impurity concentration may appear in the dependence of dT_c/dP on r, or of ΔT_c^P on r. The peculiarity in the behavior of ΔT_c^P as a function of r in the ternary solution Tl-Sb (0.084 at. %)-Hg (Fig. 4, curve 2) and in the binary solution Tl-Sb is apparently connected with this.[1]

Let us consider the cited experimental data from the point of view of the theory proposed in[5,6] which explains the presence of the maximum in the dependence of dT_c/dP on r as a function of the impurity concentration by a change in the topology of the Fermi surface under the influence of the impurity. The dependence of dT_c/dP on r for the ternary solution Tl-In-Hg (Fig. 10) is plotted in accordance with the data of Fig. 5; such dependences for the solutions Tl-Hg and Tl-In are plotted on the basis of previously published data.[1] The common feature of the dependences of dT_c/dP on r is the presence of a maximum in the solutions Tl-Hg (curve 2) and Tl-In-Hg (curve 3). According to[5,6], the variable parameter in the dependence of dT_c/dP on r is not the total residual resistance but the electron density which changes under the action of the impurity. Thus, taking into account the fact that the mercury impurity decreases the density of electron states $\nu(\epsilon_F)$, whereas an indium impurity, as has been shown previously,[4] causes it to increase, the dT_c/dP versus r curves for Tl-Hg solutions (curve 2) and of Tl-In solution (curve 3) can be represented as a single curve with a maximum (the curve ABC).

Investigating ternary systems in which one of the impurities (Sb, In) increases the electron density $\nu(\epsilon_F)$ and where the other (Hg) decreases it, one can fully observe the feature of the dependence of dT_c/dP on r which is connected with a change in the topology of the Fermi surface. Indeed the dependence of dT_c/dP on r for the Tl-In-Hg solution (curve 1) is the same as the curve ABC, only shifted towards the right on the axis of concentrations. The $\Delta T_c(r)$ dependence for the solutions Tl-Sb (0.042 at. %)-Hg and Tl-Sb (0.084 at. %)-Hg are analogous.

It has thus been established experimentally that in thallium the quantity dT_c/dP as a function of the impurity concentration has a maximum. In accordance

with the considerations developed, this corresponds to a change in the topology of the Fermi surface of thallium under the influence of the impurity (Fig. 8a). The cited data show that antimony, bismuth, and indium impurities change the Fermi energy of thallium in one direction (increasing it), whereas the mercury impurity changes it in the opposite direction (decreases it).

The topological changes of the Fermi surface manifest themselves also in the dependence of ΔT_c on the impurity concentration. If the nonlinear course of the $\Delta T_c(r)$ dependence were basically determined by a decrease of the anisotropy of the energy gap in the scattering of electrons by the impurities, then in the investigated ternary solutions the maximum change of ΔT_c should differ on increasing the impurity concentration of mercury depending on the initial content of antimony or indium impurities in the thallium (Fig. 3, curves 2–5). The maximum change of ΔT_c of ternary solutions should decrease with increasing concentration of antimony or indium impurities (Fig. 7). Actually, for the investigated ternary solutions the course of the $\Delta T_c(r)$ dependence almost does not differ from that for the binary solution Tl-Hg (Fig. 3). Consequently ΔT_c is not sensitive to the initial residual resistance of thallium and is determined by the valence of the impurity (Hg) added to the given solutions.

One can determine from the experimental data the relative change of the density of electron states in a topological transition[5] This quantity determined from two independent data on $\Delta T_c(r)$ and $\Delta T_c(P)$ should be the same. From the equality[5]

$$\Delta T_c(r) = \frac{T_c}{I_0 \nu(\varepsilon_F)} \frac{\delta\nu(\varepsilon_F - \omega_D)}{\nu_0(\varepsilon_F)},$$

where ΔT_c is the maximum change in T_c under the action of an impurity and $I_0\nu(\epsilon_F)$ is the tabulated value, it follows that the relative change of the density of electron states is

$$\delta\nu(\varepsilon_F - \omega_D)/\nu_0(\varepsilon_F) = 2.3\%.$$

The same quantity can be determined from the experimental data on the $\Delta T_c(P)$ dependence taking into account the fact that the nonlinear component of ΔT_c^n, whose maximum value is ~ 0.2[2], is responsible for topological changes. Hence

$$\delta\nu(\varepsilon_F - \omega_D)/\nu_0(\varepsilon_F) = 2.6\%.$$

According to the theory of Makarov and Bar'yakhtar[5]

$$\delta\nu(\varepsilon_F - \omega_D)/\nu_0(\varepsilon_F) \sim (\omega_D/\varepsilon_F)^{1/2} \approx 3.2\%.$$

Thus the entire aggregate of the experimental data on thallium [$dT_c/dP(r)$ and $\Delta T_c(P)$] is described by a theory taking into account the change in the topology of the Fermi surface under the influence of impurities and pressure.[5,6] In the case of thallium this apparently corresponds to the fact that under the influence of a mercury impurity the part of the Fermi surface located in the fifth or sixth Brillouin zone disappears.[14] On the other hand, the changes of T_c and dT_c/dP as a function of r, connected with a change in the anisotropy of the energy gap under the influence of impurities and of the pressure, are not decisive.

The authors consider it their duty to thank V. G. Bar'yakhtar and B. G. Lazarev for a useful discussion of the obtained results.

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