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EFFECT OF IMPURITIES ON THE TOPOLOGY OF THE FERMI SURFACE OF INDIUM

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Binary and ternary In solid solutions with Sn, Pb, Cd or Hg are studied. The quantity dT_K/dP as a function of the impurity concentration is found to have a maximum. This type of dependence indicates that the topology of the Fermi surface in indium is modified by the impurity.

IN the investigation of the joint influence of an impurity and pressure on the temperature of the Tl superconducting transition, it was observed that the derivative dT_c/dP of the temperature of the superconducting transition with respect to the pressure has a maximum as the function of the impurity concentration C and of the pressure.[1,2] According to the developed theory, this phenomenon is connected with the change of the topology of the Fermi surface under the influence of the impurity and the pressure[3]. In the case of Tl, the nonlinear dependence of dT_c/dP on the pressure and the impurity concentration is connected with the occurrence of a small Fermi-surface cavity in the sixth Brillouin zone[2,3].

In a preliminary communication[4] reporting an investigation of the influence of a Cd impurity on dT_c/dP in In, a minimum was likewise observed in the dependence of dT_c/dP on C, thus evidencing a change of the topology of the Fermi surface of In under the influence of the Cd impurity. It was recently observed that dT_c/dP of Re has a maximum as a function of the impurity concentration and of the pressure[5]. The authors likewise connected this behavior of dT_c/dP with a change in the topology of the Fermi surface of Re.

According to present day notions concerning the shape of the Fermi surface, a change in the topology of the Fermi surface under the influence of impurities and pressure can be observed in metals other than those indicated above (for example in Cd, Zn, Al). A study of the joint influence of the impurities and pressure on T_c of metals can serve as a method for observing changes in the topology of their Fermi surfaces[2]. This communication is devoted to a further investigation of the influence of impurities with different valences on the topology of Fermi surface of In.

SAMPLES AND MEASUREMENT METHOD

We investigated solid solutions of In with impurities of Cd, Hg, Sn, Pb (binary solutions), and also the ternary solution In-Pb (2 at%)-Cd, in which the Cd concentration ranged from 0 to 4%.[1)]

The indium with the impurities were placed in an ampoule made of pyrex glass. The ampoule was evacuated to a pressure $(1-2) \times 10^{-5}$ mm Hg and was sealed off at the same vacuum, after which it was placed in a furnace, where the alloy was kept for 5—7 days at a temperature slightly exceeding the melting temperature, and was stirred periodically. The alloy was then cooled abruptly. The alloys with small concentrations were made by diluting alloys of higher concentrations.

The samples were produced in the forms of wires 0.4 mm in diameter and 10—15 mm long, by pressing through a die, after which they were annealed for 7—12 days, and in some cases for several months at 100—130°C, depending on the alloy. Sufficiently good samples with a homogeneous impurity distribution were obtained, as could be judged from the width of the superconducting transition.

The ternary alloys In—Pb (2%)-Cd were prepared in the following manner. At first alloys of maximum concentration In-Pb (2%)-Cd (4%) and In-Pb (2%) were prepared. Ternary alloys with smaller Cd concentrations were obtained by diluting the maximum-concentration alloy with the binary In-Pb (2%) solution. From the x-ray diffraction investigations (Fig. 2) and from the width of the superconducting transition (Fig. 1) it can be concluded that the ternary solutions are single-phase within the range of the investigated concentrations.

[1)]The concentrations are given in atomic percent throughout.

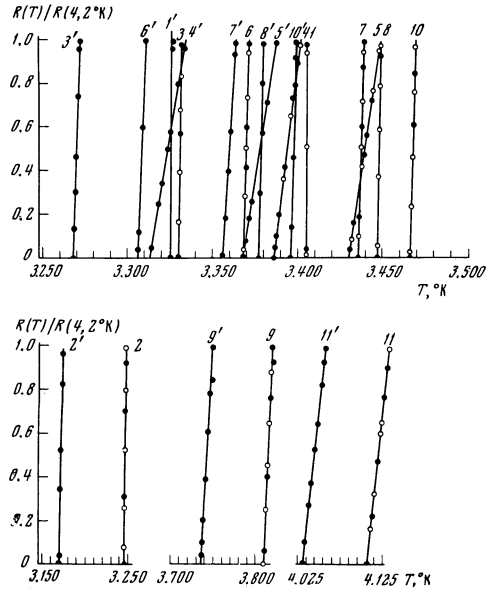

FIG. 1. Temperature dependence of R(T)/R (4.2° K): curves 1, 1′–pure In; 2, 2′–In-Cd (3.7 at. %); 3,3′–In-Cd (1.3 at. %; 4, 4′–In-Pb (2 at. %)–Cd (4 at. %); 5, 5′–In-Pb (2 at. %)–Cd (2, 3 at. %); 6, 6′–In-Hg (0.8 at. %); 7, 7′–In-Hg (4.5 at. %); 8, 8′–In-Sn (1.1 at. %); 9, 9′–In-Sn (4.5 at. %); 10, 10′–In-Pb (2 at. %); 11, 11′–In-Pb (8 at. %). curves 1–11 were plotted at P = O, and 1′–11′ at P = 1730 atm.

The alloys were investigated by the Debye method in Ti radiation. The parameters a and c of the tetragonal lattice were determined from the (131) and (222) lines accurate to $\pm 1 \times 10^{-3}$ Å. The plot of the parameters of the In-Pb (2%) lattice against the Cd impurity concentration shown in Fig. 2[2)] practically coincides with a similar plot for the binary solution In-Cd[6]. The temperature of the superconducting transition was determined from the change of the resistance ratio R(T)/R(4.2°K) with temperature (Fig. 1).

To determine the derivative of the temperature of the superconducting transition with respect to the pressure, dT_c/dP, we employed an "ice technique" using water and water-alcohol solutions[7,8]. As before[9], the shift of T_c of pure In at P = 1730 atm amounted to 80×10^{-3}°K ($dT_c/dP = -4.62 \times 10^{-5}$ deg/atm). The temperature of the superconducting transition of the indium alloys decreased linearly with pressure in the interval 0–1730 atmospheres. This made it possible to determine the change of dT_c/dP of the alloys from the shift of T_c at a constant pressure, usually 1730 atm.

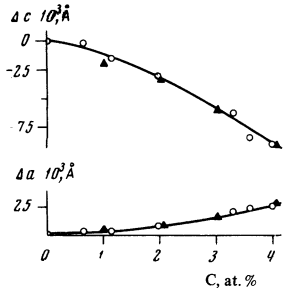

FIG. 2. Change of lattice parameters of In-Pb (2 at.%) and pure In following introduction of the Cd impurity: O–In-Cd, Δ–In-Pb (2 at.%)–Cd.

[2)]The authors consider it their duty to thank L. F. Belyak for x-ray diffraction investigations of the ternary solutions.

The measurements were made in the following manner. Four samples were placed simultaneously in the high-pressure bomb; three were alloys and one was pure In serving as a pressure indicator. The shift of the superconducting-transition temperature of the alloy samples with and without pressure was determined relative to the pure In in a single experiment. Under these conditions, there was maintained a good homogeneity of the pressure (± 20 atm), which was determined beforehand, likewise using four samples of pure In placed in the bomb. The temperature was determined with an oil manometer accurate to $(1-2) \times 10^{-3}$ deg. The width of the superconducting transition of the samples under pressure remained practically unchanged (Fig. 1). The error in the measurements of dT_c/dP amounted to $\pm 0.10 \times 10^{-5}$ deg/atm.

MEASUREMENT RESULTS

Judging from the form of the dependence of dT_c/dP on the impurity concentration, binary solutions can be divided into two groups: solutions with impurities whose valence is larger than that of In, namely In-Pb and In-Sn, and solutions with impurities whose valence is lower than that of In, namely In-Cd and In-Hg.

1. In-Pb and In-Sn solutions

Indium with Pb or Sn as an impurity exhibits a nonlinear dependence of dT_c/dP on the impurity concentration (Fig. 3a). It is seen from the figure that when the impurity Pb or Sn is added to the pure In, the absolute value of dT_c/dP decreases, reaching at a concentration of 2.4% Pb and 1.2% Sn a value 4.1×10^{-5} deg/atm. With further increase of the Pb or Sn concentration, the absolute value of dT_c/dP increases. A nonlinear dependence of dT_c/dP on the impurity concentration is observed for the In-Pb solution in a wider range of concentrations than in the In-Sn solution.

The dependence of the superconducting-transition temperature shift ΔT_c of In on the impurity concentration is the same for both the Pb and Sn impurities (Fig. 3d): at low Pb or Sn concentrations the temperature of the superconducting transition decreases very slightly; an appreciable growth of the superconducting-transition temperature is observed with further increase of the impurity concentration. The presented experimental data (Fig. 3b) agree with previously published data by others[10,11].

2. In-Cd and In-Hg Solutions

In these solutions, the dependence of dT_c/dP on the concentration of the impurity is also nonlinear (Fig. 4a)[4,12]. The value of dT_c/dP at the maximum, for both In-Cd and In-Hg solutions, is the same and equals -354×10^{-5} deg/atm at 1.3% Cd and 0.8% Hg. The dependence of the shift ΔT_c on the impurity concentration differs greatly for the In-Hg solution from that for the In-Cd solution (Fig. 4b).

3. In-Pb (2%)-Cd Solution

It is seen from Figs. 3a and 4a that the maximum values of dT_c/dP are different for the solutions In-Pb and In-Sn, from those for In-Cd and In-Hg. To explain the nature of this difference, we have investigated

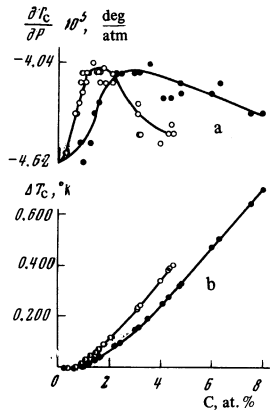

FIG. 3

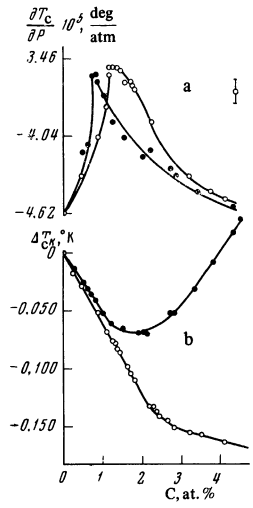

FIG. 4

FIG. 3. Plots of $\partial T_c/\partial P$ (a) and ΔT_c (b) of In vs. the impurity concentration: ●—In-Pb, ○—In-Sn.

FIG. 4. Plots of $\partial T_c/\partial P$ (a) and ΔT_c (b) of In vs. the impurity concentration: ○—In-Cd, ●—In-Hg.

In-Pb (2%)-Cd ternary solutions. The dependence of dT_c/dP on C for these solutions (Fig. 5a) is more complicated than for binary solutions (Figs. 3a and 4a). Addition of the Cd impurity (up to ~1.5%) to the In-Pb (2%) solution leads to a decrease of the value of $dT_c/dP = -4.1 \times 10^{-5}$ deg/atm of the solution to a value corresponding to pure In (-4.62×10^{-5} deg/atm). With further increase of the Cd impurity concentration, the dependence of dT_c/dP on C is similar to that for the In-Cd solution.

The ΔT_c(C) dependence of the In-Pb (2%)-Cd solutions is shown in Fig. 5b. The superconducting-transition temperature of the initial In-Pb (2%) solution decreases slightly when up to 1.8% of Cd is added, and with further increase of the Cd concentration there is observed a more rapid decrease of T_c of the In-Pb (2%)-Cd solution. The change of the superconducting-transition temperature in this system amounts to 85 $\times 10^{-3}$ deg.

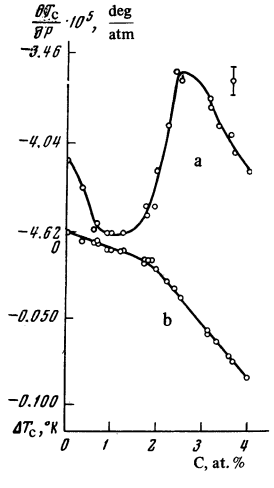

FIG. 5. Plots of $\partial T_c/\partial P$ (a) and ΔT_c (b) in the In-Pb (2 at. %) system vs. the concentration of the Cd impurity.

DISCUSSION OF RESULTS

1. Nonlinear Dependence of dT_c/dP on the Impurity Concentration

According to the developed concepts, the nonlinear dependence of dT_c/dP on the impurity concentration C (Figs. 3a, 4a, 5a) is connected with the change of the topology of the Fermi surface under the influence of the impurity[3]. The character of the topological transition can be evaluated from the asymmetry of the $dT_c/dP = f(C)$ curve[3], by using Fig. 6 and assuming that introduction of the impurity leads to a change in the Fermi energy.

If it is assumed that the impurities whose valence is larger than that of In change the Fermi energy in one direction (for example, they increase it), and the impurity whose valence is smaller than that of In changes it in the opposite direction (for example, decrease), then the scale of the concentration characterizes the change of the Fermi energy of the pure In under the influence of the impurity. Then, from a comparison of the experimental curves (Figs. 3a and 4a) with the theoretical ones (Figs. 6a and 6b) we can draw the following conclusion: When the Fermi energy increases (In-Pb and In-Sn alloys, Fig. 3a) a cavity of the Fermi surface is produced (Fig. 6b), and when the Fermi energy decreases (alloys In-Hg and In-Cd) (Fig. 4a) breaking of the "neck" occurs (Fig. 6a, $\epsilon_c < \epsilon_F$).

The observed maximum of dT_c/dP in In-Sn and In-Pb solutions and also in In-Hg and In-Cd solutions at different impurity concentrations can be attributed to the different effective action of the impurity on the density of the electron states. Indeed, if it is assumed that the efficiency of the Sn impurity is equal to unity ($C_{eff} = C_{Sn}$), and that of Pb is equal to 0.5 ($C_{eff} = 2C_{Pb}$) in the In-Sn and In-Pb solutions, then the dependence of dT_c/dP of these solutions on the effective impurity concentration is described by a single curve (Fig. 7a). A similar construction can be made for the solutions In-Hg and In-Cd (Fig. 7b, $C_{eff} = C_{Hg} = 1.67C_{Cd}$). The fact that the singularities in the solutions In-Hg and In-Cd differ from those in In-Pb and In-Sn proves the behavior of dT_c/dP as a function of the concentration of the Cd impurity in the In-Pb (2%)-Cd solutions (Fig. 5a). The Cd impurity (~1.5%) eliminates the changes of dT_c/dP due to the impurity Pb (2%), and the topology of the Fermi surface of the In-Pb (2%)-Cd (1.5%) solution corresponds to the topology of the Fermi surface of pure In. With further increase of the Cd impurity, the dependence of dT_c/dP on C of the In-Pb (2%)-Cd solution is

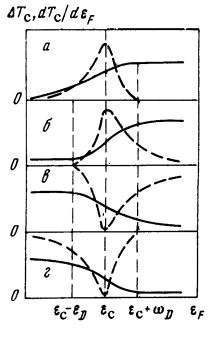

FIG. 6. Plot of T_c (solid curve) and of $dT_c/d\epsilon_F$ (dashed) vs. the Fermi energy in the simplest topological cases: a—formation of a Fermi-surface neck if $\epsilon_c < \epsilon_F$ or a break in the neck if $\epsilon_c > \epsilon_F$ ($dT_c/d\epsilon_F > 0$); b—formation of one of the parts of the Fermi surface if $\epsilon_c > \epsilon_F$ ($dT_c/d\epsilon_F > 0$); c—vanishing of the neck of the Fermi surface if $\epsilon_c > \epsilon_F$, or formation of one if $\epsilon_c < \epsilon_F$ ($dT_c/d\epsilon_F < 0$); d—vanishing of one of the parts of the Fermi surface if $\epsilon_c > \epsilon_F$ ($dT_c/d\epsilon_F < 9$). Here ϵ_c—critical energy at which a change in the topology of the Fermi surface takes place, and ω_D—Debye temperature.

similar to the dependence of the In-Cd solution (Figs. 4a and 5a).

The nonlinear dependence of dT_c/dP of the ternary solution In-Pb (2%)-Cd shows indeed that an impurity whose valence is lower than that of In changes ϵ_F in one direction, and an impurity whose valence is larger than that of In changes it in the opposite direction. We can then construct for In the change of dT_c/dP ($\Delta dT_c/dP$) due to the change of the Fermi surface and to the change of the electron density $\Delta n = \alpha C \Delta z$ (Fig. 8a). Here Δz is the difference between the valence of the impurity atom and the host-metal atom, and α is a positive constant on the order of unity.

It was already noted (Figs. 3a, 4a, 5a, 7a, 7b) that 1 at.% of a different impurity changes ϵ_F or Δn by a different amount. These results make it possible to establish a unified scale of $\Delta n = \alpha C^*_{eff} \Delta z$ for both $\Delta n > 0$ and $\Delta n < 0$. Thus, 1 at.% of Hg impurity causes the same absolute change in the Fermi energy or in the electron density as does 1.67 at.% of Cd or 1.1 at.% Sn, or 2.2 at.% Pb ($C^*_{eff} = C_{Hg} = 1.67C_{Cd} = 1.1C_{Sn} = 2.2C_{Pb}$).

The maximum of the quantity dT_c/dP corresponds to the fact that the Fermi energy coincides with the critical energy ϵ_c at which a change takes place in the Fermi-surface topology (Figs. 6a and 6b). From a comparison of the curves of Figs. 6a and 6b and of Fig. 8a we can conclude that the difference between the critical energies that are connected with the change of the topology of the Fermi surface under the influence of the impurities whose valence is smaller than that of In, $\epsilon^{(2)}$, and of the impurities whose valence is larger than that of In, $\epsilon^{(1)}$, remains not less than $2\omega_D$, which is of the order of several hundredths of an electron volt.

As is well known, the density of the electronic states

$$\nu(\varepsilon) = \int_{\varepsilon} dS/|v|,$$

where $\nu = \partial\epsilon/\partial p$ and dS is an element of the surface $\epsilon = \epsilon(p)$, has singularities at those critical values of the energy $\epsilon = \epsilon_c$ at which the topology of the surface $\epsilon(p) = \epsilon_F$ changes[13]. Using the data of Fig. 8a, we can construct the $\nu(\epsilon)$ dependence for In near the Fermi energy (Fig. 8b).

These singularities should become manifest also in the electronic properties of the metal in the normal state[14], for example, in the dependence of the electronic heat capacity of In on the impurity concentration, or in the dependence of the electronic coefficient of thermal expansion on the impurity concentration[3)].

The experimental data on the dependence of the magnetic susceptibility of In on the impurity concentration and on the temperature[15] can apparently be attributed to the presented $\nu(\epsilon)$ dependence of the magnetic susceptibility on the concentration of the Cd impurity at C = 2% was connected with the passage through a local energy minimum located in the third Brillouin zone near the W point[15]. This is accompanied by breaking of the "necks" of the β tubes of the Fermi surface of In, located in the third Brillouin zone. A similar change in the topology of the Fermi surface follows from the non-

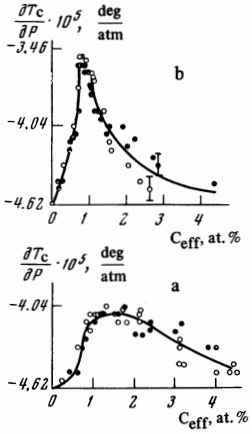

FIG. 7.

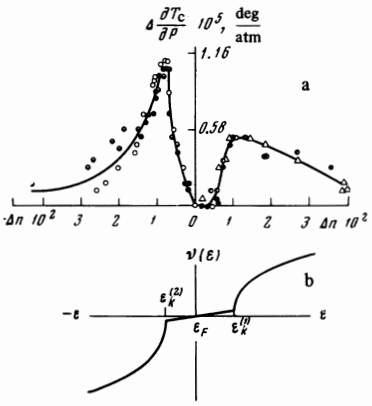

FIG. 8

FIG. 7. Plot of $\partial T_c/\partial P$ vs. the effective impurity concentration: a—for In in the following systems: ○—In-Sn, ●—In-Pb ($C_{eff} = C_{Sn} = 2C_{Pb}$), and b—for In and In-Pb (2 at.%) in the following systems: ●—In-Cd, ○—In-Hg, ◓—In-Pb (2 at.%)—Cd ($C_{eff} = C_{Hg} = 1.67C_{Cd}$).

FIG. 8. a—Plot of $\partial T_c/\partial P$ of In vs. $\Delta n = \alpha C^*_{eff} \Delta z$. b—Density of electronic states of In near the Fermi energy ϵ_F ($\epsilon_c^{(1)}$, $\epsilon_c^{(2)}$—critical energies at which a change takes place in the topology of the Fermi surface.

linear dependence of dT_c/dP on C in the In-Cd and In-Hg solutions (Figs. 4a, 7b, 8a).

When an impurity whose valence is larger than that of In (Pb or Sn) is added, the magnetic susceptibility χ decreases monotonically with the concentration[15]. However, the temperature dependence $\chi(T)$ reveals a reversal of the sign of $d\chi/dT$ at 3—7% Pb[15]. The change of the sign of $d\chi/dT$ in the In-Pb solution was attributed to the occurrence of a small part of the Fermi surface located in the fourth Brillouin zone[15]. From a comparison of the experimental data (Fig. 3a, 7a, 8a) with theory (Fig. 6b) it also follows that a Pb or Sn impurity causes formation of a part of the Fermi surface that may be located in the fourth Brillouin zone, but one cannot exclude the possibility of formation of Fermi-surface β tubes located in the third Brillouin zone, which exists in accordance with the theory proposed in[16], but which were not observed in a number of experiments[17].

Singularities in the dependence of χ on the impurity concentration and on the temperature were observed also in other metals with impurities[17,18]. Thus, in the case of Bi with Pb or Sn impurity there is observed a maximum in the $\chi(C)$ dependence[18], and in the case of Al with Mg impurity the sign of $d\chi/dT$ reverses at 3.5 at.%[19]. From an investigation of the Fermi surface of Bi and Al with impurities, based on the de Haas—van Alphen effect, it follows that in the case of Bi with impurities, at concentrations corresponding to the maximum in the $\chi(C)$ dependence, there vanishes a group of electrons[20], and in the case of Al with 3.3 at.% Mg, there possibly occurs a change in the topology of the Fermi surface[21].

Thus, the presented experimental data (Figs. 3a, 4a, 5a), and also the experimental data on Tl and Re, show that insignificant changes of the Fermi surface topology exerts a great influence on the value of dT_c/dP as a

[3)] Unfortunately, no such experiments have been performed as yet.

function of the impurity concentration and of the pressure.

2. Dependence of Superconducting-transition Temperature on the Impurity Concentration

The $T_c(C)$ dependence for a number of metals is nonlinear[22]. This is connected with the fact that besides the linear mechanism there exists also a nonlinear mechanism in the $T_c(C)$ variation. The linear mechanism is usually connected with the linear changes of the Debye temperature, of the electron-phonon interaction constant, and of the density of the electronic states under the influence of the impurity. The nonlinear mechanism in the $\Delta T_c(C)$ dependence is connected with the decrease of the influence of the anisotropy of the energy gap in the spectrum of the conduction electrons[22] and the possible change of the Fermi-surface topology under the influence of an impurity[3]. These mechanisms are different in character.

Indeed, when a change occurs in the Fermi-surface topology, the nonlinear change of $\Delta T_c(C)$ is connected with a singularity in the density of the electronic states (Fig. 6). In this case, too, the experimental dependence $\Delta T_c(C)$ can be regarded as a sum of a linear and nonlinear mechanisms. When identical topological changes occur as a result of different impurities, the experimental $\Delta T_c(C)$ curves differ only in the linear mechanism. To verify this it is necessary to take the difference between the experimental curves. However, it is necessary to take into account here the different change of $\nu(\epsilon)$ under the influence of the impurities, by introducing C_{eff} (which is obtained by aligning $\partial T_c/\partial P = f(C)$ as shown above).

Another possible nonlinear mechanism in $\Delta T_c(C)$ is connected with the smearing of the anisotropic part of the electron-electron interaction under the influence of the impurity[22]. Just as in the case of the nonlinear mechanism considered above when the experimental curves are plotted in coordinates that take into account the individuality of the impurities, they differ only because of the linear mechanisms. The individuality of the impurity is connected with the anisotropy of the scattering center and is characterized by the parameter λ_i[22]. The relative difference between the impurities is determined by comparing the dependences of the ratio $\Delta T_c/C$ on $\ln C$[22]. On the basis of this theory, explanations were obtained for the experimental $\Delta T_c(C)$ dependences of the systems In-Hg, In-Cd, In-Pb, and In-Sn[22] (Fig. 9, curves 1 and 2). Using the data obtained in the study of binary systems, we can construct a theoretical curve for $\Delta T_c(C)$ of the ternary In-Pb (2%)-Cd alloys, since the nonlinear mechanism depends only on the amount of the impurity (residual resistance), while the linear is the same as in the In-Cd alloy. From Fig. 9 (curves 3 and 4) we see that there is a qualitative difference between the theoretical and experimental curves. Taking into account the nonlinear dependence of $\partial T_c/\partial P$ on C of this system, it is natural to attribute the indicated difference to the existence of another nonlinear mechanism connected with the change of the Fermi-surface topology. The fact that the nonlinear mechanism is distinctly manifest in ternary systems is connected with the fact that the magnitude of the anisotropy mechanism is smaller here than in binary systems (Fig. 9, curves 5 and 4″).

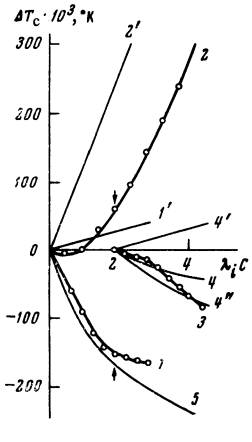

FIG. 9. ΔT_c vs. $\lambda_i C$ of indium alloys. Thick curves—experimental: 1—In-Cd, 2—In-Pb, 3—In-Pb(2 at.%)—Cd; thin curves—constructed in accordance with the theory of [22]: 5—nonlinear mechanism of $T_c(C)$ dependence of indium alloys ($\lambda = 1 = \lambda_{Sn}$, $\lambda_{Cd} = 0.63$, $\lambda_{Pb} = 0.935$); 1′, 2′—linear mechanism in the case of the In-Cd and In-Pb systems, 4—curve for In-Pb(2 at.%)—Cd, representing the sum of the linear mechanism (curve 4′) and the nonlinear one (curve 4″). The arrows designate the points corresponding to 2 at.% Pb.

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

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1