Electron-phonon interaction in indium-magnesium alloys with a structural phase transition

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$In_{1-x}Mg_x$ alloys in the face-centered tetragonal (FCT) phase are investigated by the electron single-particle tunneling technique. It is shown that the electron-phonon coupling constant λ increases as the FCT⇄FCC (face-centered cubic) phase transition point x_m is approached. The increase in λ is due both to a growth in the electron state density at the Fermi level $\nu(\varepsilon_F)$ and to the redistribution of phonon frequencies that is observed on the basis of the spectral functions of the electron-phonon interaction $g(\omega)$. It is suggested that the growth of $\nu(\varepsilon_F)$ and the deformation of $g(\omega)$ are due to the Lifshitz electron transition which occurs in these alloys at $x_l \approx 0.02$.

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1. INTRODUCTION

In the system of alloys $In_{1-x}Mg_x$, where x is the impurity concentration, a structural phase transition (SPT) takes place at $x = x_m = 0.04$, from the face-centered tetragonal (FCT) to the face-centered cubic (FCC) phase. In the FCT phase of these alloys, the axial ratio c/a (a and c are the parameters of the crystal lattice) changes with increase in the magnesium content from 1.076 at $x = 0$ to 1.04 at x_m. At higher concentrations of magnesium, the degree of tetragonality falls off discontinuously to unity, and at $x > 0.06$ the alloy has a FCC lattice,[1] see Fig. 1, curve 1.

The Fermi surface of pure indium in the third Brillouin zone consists of so-called "β rings," which lie in planes perpendicular to the c axis.[3] The absence of "α tubes" between the points W and T is associated with the tetragonal distortion of the crystal lattice. Therefore we can assume that the decrease in the degree of tetragonality in the case of an increase in the magnesium concentration should lead to a topological transition in the Fermi surface, namely, to the appearance of embryo of an α tube in the third Brillouin zone.

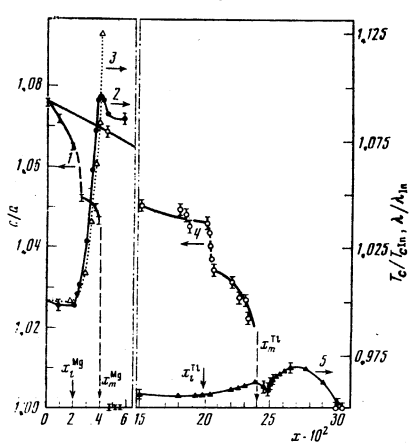

FIG. 1. Concentration dependences of the axial ratio c/a and of T_c/T_{cIn} for the face-centered tetragonal phase of alloys of indium with magnesium (curves 1 and 2) and with thallium (curves 4 and 5); curve 3—the dependence of the electron-photon interaction constant λ (referred to λ_{In}) on the magnesium concentration in the neighborhood of a structural phase transition.

Usually, the addition of an impurity of lower valence (decrease in the number of electrons per atom) leads to a drop in the critical temperature of the superconducting transition T_c and therefore the sharp increase in T_c in the region of magnesium concentration $x > 0.02$ (Fig. 1, curve 2) is at first glance unexpected.[3] This unusual behavior of the critical temperature, which precedes the structural phase transition, indicates possible peculiarities in the electron-phonon interaction in this region of magnesium concentration.

In contrast to the systems In-Cd and In-Tl, in which phase transitions similar to FCT ⇄ FCC have also been observed (Ref. 4 and citations given there), the system of In-Mg alloys has been studied in less detail.

In the present work, we give the results of a study of the behavior of the electron-phonon interaction in the vicinity of a structural phase transition to the tetragonal phase of indium-magnesium alloys. This study has been carried out with the help of the method of single-particle electron tunneling, which makes it possible to obtain rather complete information on electron and lattice properties of the material.

2. EXPERIMENT

1. For the preparation of the indium-magnesium alloys, metals of purity 99.999 were used. Batches of the initial materials were placed in quartz ampoules evacuated to 10^{-5} Torr. After this, the ampoules were sealed off and placed in a furnace, where the components were melted under continuous mixing. Then the alloy was rapidly cooled. Alloys of low concentrations were prepared by diluting the initial concentrated alloy.

2. Al-insulator-$In_{1-x}Mg_x$ film tunnel junctions were prepared by condensation of the metals in a vacuum of 10^{-6} Torr on a glass substrate cooled to liquid nitrogen temperatures. The film thicknesses were ~200 Å and ~3000 Å for aluminum and the alloys, respectively. The alloy films were deposited by the "flash" method at rates of ≈ 300 Å/sec. The regime of oxidation of the aluminum film to produce a tunnel barrier (pressure of dry oxygen ~20 Torr at a substrate temperature 350 K,

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oxidation time 30 sec) was so chosen that the resistance of the finished contacts was 20 ohms/mm$^2 \leq R_T \leq$ 200 ohms/mm^2. After heating, the film samples were annealed in high vacuum at a temperature of 310 K for not less than 10 hours. The quality of the tunnel structures was estimated from the sharpness of the peaks in the $dI(U)/dU$ curves with the contacts biased to correspond to the singularities in the energy spectra of the superconductors.

The magnesium content in the films was determined from the dependence of the critical temperature of the superconducting transition on x (Ref. 3) (Fig. 1, curve 1). The steep slope of this curve guarantees a sufficiently small error in the determination of Mg: ± 0.1 at·% in the region $0.02 \leq x \leq 0.04$. The following features of the preparation of the tunnel structures with the In-Mg alloys must be noted. The impurity densities of the initial sputtered alloy and in the final films were equal only for the portions of the films placed normal to the plane of the vaporizer, i.e., in the region of small solid angles. At large solid angles, there was no such equality. Nine substrates were placed simultaneously in the vacuum chamber at various solid angles relative to the vaporizer; this made it possible to obtain in a single evaporation several tunnel samples with different impurity densities in the alloy films. One of the reasons for this phenomenon is obviously the significant difference in the vapor pressures of indium and sublimating magnesium at the vaporization temperatures $T_{vap} \approx 1000$–1100 °C.

3. The following characteristics were recorded for each sample with the help of a high resolution tunnel spectrometer.

a) The first derivative of the tunnel current with respect to the voltage $\sigma(U) = dI(U)/dU$ is the differential conductivity, from which the superconducting gap of the alloy Δ was determined. Examples of such characteristics for $x = 0$ and $x = 0.039$ are shown in Fig. 2, curves 1 and 2. The error in the measurements of Δ was 2×10^{-3} meV. It is seen from curve 3 of Fig. 2 that, in the region of magnesium concentrations where the rapid growth of T_c of the alloys begins, the energy gap of the superconductor also increases.

b) The critical temperature T_c of the superconducting transition of the alloys was determined by the "tunnel" method—by observing the appearance of a gap of the superconductor (in the case of a decrease in the temperature, the resistance of the tunnel contact increases at $U = 0$ at the instant of appearance of the gap).[5] Such a method makes it possible to measure the T_c of the small portion of the alloy film that is located directly in the tunnel contact with the injector. The experimental error amounts to 5×10^{-3} K.

c) The second derivative $d^2I(U)/dU^2$, from which were found the characteristic phonon frequencies ω_i of the spectral function of the gap $\Delta(\omega)$ which reflects the Van Hove singularities of the phonon density of states $F(\omega)$.[6] Typical characteristics of this type are shown in Fig. 3, curves 1 and 2. The error in the determination of ω_i amounted to 3×10^{-2} meV and 5×10^{-2} meV for the frequencies ω_t and ω_l, respectively. It is seen from Fig. 3 that the frequencies of the critical point of the transverse mode (ω_{t1}) and of the Van Hove singularities of the longitudinal mode of oscillations of the phonon spectrum (ω_l) decrease by an amount ~3% as $x \to x_m$ (curves 3 and 5). The frequency ω_{t2} increases insignificantly as $x \to x_m$ (curve 4).

d) The differential resistances of the contacts $R(U) = dU/dI$ in the superconducting $R_S(U)$ and normal $R_N(u)$ states. These data serve as a basis for finding the real and imaginary parts of the function $\chi(\omega) = \omega[\omega^2 - \Delta^2(\omega)]^{-1/2}$, $\omega \equiv U$, which are necessary for the determination of the tunnel density of states $N_T(\omega) = R_N/R_S$ and the energy dependence of the gap $\Delta(\omega)$.[7]

All the measurements were carried out at a temperature 1.4 K. Indium-magnesium alloys of twenty different concentrations were investigated in the given research; 2–3 samples of the same concentration were prepared and measured, while measurement of the characteristics mentioned above were repeated not less than three times for each sample.

4. The spectral function $g(\omega) = \alpha^2 F(\omega)$ of electron-phonon interaction, the electron-phonon interaction con-

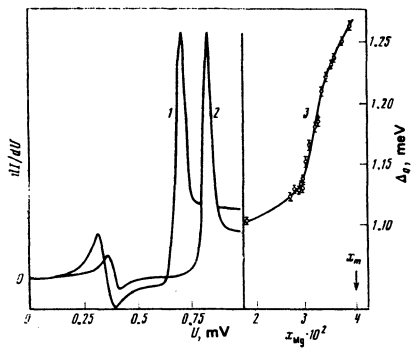

FIG. 2. First derivatives of the volt-ampere characteristics (VAC) of tunnel samples based on $In_{1-x}Mg_x$; curve 1—$x = 0$, curve 2—$x = 0.039$. Curve 3—dependence of the gap in the energy spectrum of superconducting In-Mg alloys on the magnesium concentration.

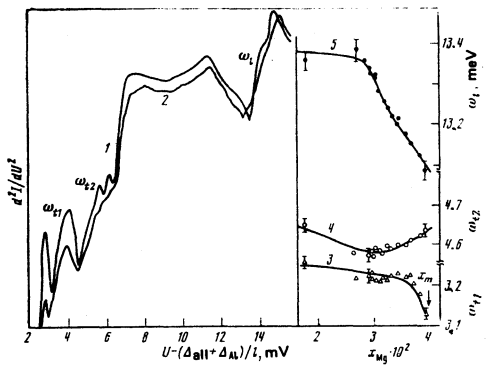

FIG. 3. Second derivatives of the VAC tunnel samples based on $In_{1-x}Mg_x$; curve 1—$x = 0$, curve 2—$x = 0.039$. Curves 3, 4, 5 are the concentration dependences of the Van Hove singularities ω_{t1}, ω_{t2} and ω_{t3}, respectively, of the phonon spectrum ω_i of the In-Mg alloys in the FCT phase as $x \to x_m$; Δ_{all} is the gap in the energy spectrum of the alloy.

stants

$$\lambda=2\int_0^\infty \omega^{-1}g(\omega)d\omega,$$

the Coulomb pseudopotentials μ^*, the electron parameters $\bar{E}$, the mean values over the frequency spectrum $\langle\omega\rangle$ and $\langle\omega^2\rangle^{1/2}$, the quantities A^2, and also the calculated values T_c^{calc} were calculated by the method of Ref. 7, using the dispersion relation for the real and imaginary parts of the function $\chi(\omega)$, see Fig. 4. The expressions determining the enumerated quantities, and the results of their calculations for pure indium and five different alloys, are given in the Table. Each of the quantities in the Table is an average for three samples with the same concentration. The following notation is used in the Table:

$$A^2=\int_0^\infty \alpha^2(\omega)F(\omega)d\omega; \quad \bar{E}=\int_0^\infty \omega\alpha^2(\omega)F(\omega)d\omega;$$

$$\langle\omega\rangle=2\lambda^{-1}\int_0^\infty \alpha^2(\omega)F(\omega)d\omega; \quad \langle\omega^2\rangle=2\lambda^{-1}\int_0^\infty \omega\alpha^2(\omega)F(\omega)d\omega;$$

$$\mu^*=\frac{\nu(\varepsilon_F)U_c}{1+\nu(\varepsilon_F)U_c\ln(\varepsilon_F/\omega_D)},$$

where U_c is the screened Coulomb potential, $\nu(\varepsilon_F)$ is the density of electron states at the Fermi level and ω_D is the Debye energy.

An increase in concentration of magnesium leads to a deformation of the $g(\omega)$ curves: to a shift down in energy, to an increase in the amplitude of the peak corresponding to transverse phonons, and to a smearing and a lowering of the peak of the longitudinal mode of the oscillations (Fig. 5).

3. DISCUSSION OF RESULTS

It is seen from the results of tunnel experiments with the alloys $In_{1-x}Mg_x$ shown in Fig. 1 (curve 3) that, as the composition approaches the phase equilibrium boundary x_m, the electron-phonon interaction increases, the constant λ at the bounding alloy with $x=0.039$ is 12% greater than in pure indium. This fact furnishes a natural explanation for the increase in the critical temperature of the alloys T_c with $x>0.02$ (see the Table).

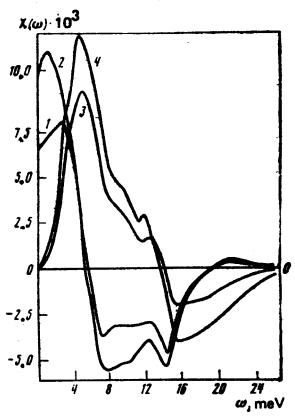

FIG. 4. Tunnel density of states (curves 1 and 2) and dispersion integral (curves 3 and 4) of indium and of the alloy with $x=0.039$.

TABLE I.

$x_{Mg}\cdot10^2$,	T_c^{exp}, K	2Δ, meV	$2\Delta_0/T_c$	λ	μ^*	A^2, meV	$\bar{E}=\frac{\nu(\varepsilon_F)\langle I^2\rangle}{2M}$, meV^2	$\langle\omega\rangle$, meV	$\langle\omega^2\rangle^{1/2}$, meV	T_c^{calc}, K
0	3,407	1,106	3,775	0,80	0,120	2,78	25,02	6,95	7,9	3,39
2,7	3,456	1,136	3,822	0,81	0,104	2,77	24,20	6,83	7,73	3,449
3,0	3,53	1,168	3,850	0,83	0,115	2,80	24,36	6,75	7,66	3,521
3,4	3,666	1,222	3,876	0,854	0,120	2,86	24,80	6,70	7,62	3,653
3,6	3,674	1,235	3,909	0,867	0,110	2,86	25,00	6,60	7,59	3,663
3,9	3,732	1,259	3,923	0,91	0,123	2,89	25,60	6,35	7,50	3,721

We estimate the contributions from the electron and phonon subsystems to the coupling constant λ of the studied alloys in the range of concentrations $0.02\leq x\leq 0.04$. For this purpose, we use the empirical formula of MacMillan, differentiated with respect to x:

$$\lambda=\nu(\varepsilon_F)\langle I^2\rangle/M\langle\omega^2\rangle=2\bar{E}/\langle\omega^2\rangle,$$

where $\nu(\varepsilon_F)$ is the density of electron states on the Fermi surface, I is the matrix element of electron-ion interaction, M is the average mass of an atom of the alloy:

$$\frac{d\ln\lambda}{dx}=\frac{d\ln\bar{E}}{dx}-2\frac{d\ln\langle\omega^2\rangle^{1/2}}{dx}. \qquad (1)$$

Since the electron-phonon interaction constant does not depend on the average mass of an atom of the alloy,[8] the first term in (1) is equal to

$$\frac{d\ln\bar{E}}{dx}\approx\frac{d\ln[\nu(\varepsilon_F)\langle I^2\rangle]}{dx}$$

and represents the purely electron contribution to λ. A numerical estimate (the values of all the parameters are taken from the Table) shows that the contributions of the electron and phonon terms are of the same order of magnitude and together insure an increase in λ. It is well known that the electron-phonon interaction constant should decrease with decrease in the valence.[8] The opposite effect is observed in the system that we have investigated. This fact compels us to assume that a rearrangement takes place in the electron spectrum of the alloys $In_{1-x}Mg_x$ with increase in concentration, at the value $x_1\approx 0.02$, bringing with it an increase in the den-

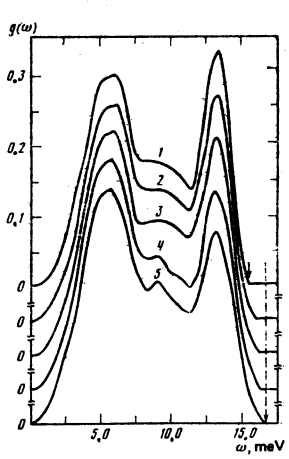

FIG. 5. Spectral functions of the electron-photon interaction for different $In_{1-x}Mg_x$ alloys in the FCT phase: curves 1–5 correspond to the concentrations $x=0$, 0.027, 0.034, 0.036, and 0.039.

sity of electron states [and of the quantity $\nu(\varepsilon_F)\langle I^2\rangle$], namely, a phase change of $2\frac{1}{2}$ order (the Lifshitz transition).[9]

Actually, as has already been noted in Sec. 1, nucleation can taken place in indium alloys that have impurities with $Z < Z_{In} = 3$, because of the change in the axial ratio of the lattice parameters c/a; as $x \to x_m$, an increase occurs in the cross section of the α tube in the third Brillouin zone (Ref. 2).[1)] It follows from this assumption that the Fermi surface of the In-Mg alloys in the FCC phase (γ phase) i.e., as $x \to x_m$ it is similar to the Fermi surface of aluminum. In order to verify this, planar cross sections of the Fermi surface were constructed for an alloy with $x = 0.065$ of the FCC phase ($a = 4.693$ Å, $Z = 2.935$, Ref. 1) with the help of the Harrison procedure and the validity of this assertion in the free electron model was demonstrated.

2. The following considerations support the validity of the assumption as to the topological electron transition: assuming no rearrangement of the electron spectrum, we shall attribute the change in the density of electron states to only a decrease in the volume Ω of the unit cell of the alloy [as is known, in the free electron model, $\nu(\varepsilon_F) \propto \Omega^{-1/3}$]. Using for T_c a formula of the BCS type, we estimate the increment ΔT_c connected with $\Delta\Omega$:

$$\Delta T_c \sim T_c^* (\ln \tilde{\omega}_D / T_c^*) [1 - (\Omega_2/\Omega_1)^{1/3}].$$

Here $T_c^* = T_c(x_t)$ is the value of the critical temperature at x_t, where its increase begins. In this estimate it was assumed that the Debye frequency depends weakly on the concentration.[2)] A numerical estimate with the use of quantities corresponding to the In-Mg system [$T_c^* = T_c$ (0.02), $\tilde{\omega}_D \approx 110$ K, $\Omega_1 = 104.56$ Å,[3] $\Omega_2 = 104.16$ Å,[3] Refs. 1 and 3] gives $\Delta T_c^{est} \sim 0.015$ K; $\Delta T_c^{exp} \approx 0.4$ K is thirty times larger. Account of the dependence of T_c on the number of electrons per atom Z strengthens the result of the estimate, increasing the ratio $\Delta T_c^{exp}/\Delta T_c^{est}$. This estimate shows that it is impossible to ignore the changes in the properties of the electron subsystem in the system of alloys considered. A similar result is obtained also in the estimate of ΔT_c in the system of In-Tl alloys.[10,13] (See Fig. 1, curves 4 and 5.)

Many physical properties of alloy systems based on indium with impurities of lower or equal valency (Cd, Mg, Hg, Tl) are similar.[4,14] In particular, in all these systems, the axial ratio c/a decreases with increase of concentration in the face-centered tetragonal phase. According to the data of Ref. 10 in indium alloys in the region of mercury concentration $x \sim 0.07$–0.080 and thallium concentration $x \sim 0.2$–0.25 (we recall that at low temperatures $x_m \approx 0.28$–0.3 in this system) topological transitions are observed experimentally by the method based on the study of the irregular character of the $dT_c/dp = f(x,p)$ dependences (p is the pressure) in the vicinity of the Lifshitz transition.[11]

We note that the following experimental facts, which are in agreement with the predictions of the theory of Ref. 15, also indicate the existence of a topological transition preceding the structural phase transition in these alloys. In the set of In-Mg alloys in the region $x_t \sim 0.02$, i.e., where T_c begins to increase rapidly, there is an anomaly in the dependences of the lattice parameters, and the behavior of the relation is irregular.[1] In the system In-Tl at $x = 0.20$, too, singularities are observed in the structure, accompanied by discontinuities in the lattice parameters, in the volume of the unit cell Ω, and in the axial ratio c/a[13] (see Fig. 1, curves 1 and 4).

3. We now discuss the behavior of the phonon subsystem in the alloys $In_{1-x}Mg_x$ and its contribution to the electron-phonon interaction upon increase in the concentration of magnesium.

Upon introduction of Mg impurities in the indium matrix, local oscillations should appear in the phonon spectrum of the alloy. As follows from a simple estimate[16] which does not take into account changes in the force constants and the number of electrons per atom: $\omega_0 \sim (M/m)^{1/2}\langle\omega^2\rangle^{1/2}$ (m and M are the atomic masses of magnesium and indium, respectively), the frequency of the local oscillation of the impurity in this system is $\omega_0 \sim 15.3$ meV in order of magnitude. Account of the fact that the magnesium is an impurity of lower valence, somewhat reduces this value of the frequency ω_0. However no singularities have been observed on the $d^2I(U)/dU^2$ dependences in the 13–14 meV range [similar, for example, to the singularities in $I''(U)$ for the Pb-In system[17]], which could be connected with the local oscillations in the phonon spectrum of the alloy. It is seen from Fig. 5 that the amplitude of the peak of the longitudinal oscillations of the spectral function $g(\omega)$ falls off rapidly with increase in the concentration, while its trailing edge is smeared out with increase in concentration of the magnesium. It is not excluded that this is the result of redistribution of the frequencies in the phonon density of states because of the appearance of "local" oscillations at $\omega_t \leq \omega_0 \leq \omega_e$ (ω_e is the end-point frequency of the phonon spectrum).[3)]

As follows from Fig. 5, the character of the spectral functions of the electron-phonon interaction of the alloys changes upon increase in the concentration of the Mg impurity in the alloy: the amplitude of the transverse peak in $g(\omega)$ increases relative to the longitudinal $g(\omega_t)/g(\omega_l) = 0.91$ [for indium and $g(\omega_t)/g(\omega_l) = 1.22$ for the alloy with $x = 0.039$]. Since the transverse phonons give the greatest contribution to the electron-phonon interaction constant λ in simple polyvalent metals,[8] the observed deformation of the electron-phonon interaction is one of the reasons for the increase in the coupling constant.

Inasmuch as the discussed phase transition is not accompanied by a change in the translational symmetry, and the volume of the primitive cell changes insignificantly (i.e., the transition takes place according to a representation with $\mathbf{k} = 0$), the frequencies with small momenta should be softened; as for short-wave phonons, their rapid increase should be expected (hardening of the spectrum) in the region of a structural phase transition, since as $x \to x_m$ the volume of the elementary cell of the alloys decreases[1] (incidentally, also as a consequence of the dissolution of di-valent magnesium in the indium matrix). It is seen from the Table that the mean frequencies $\langle\omega\rangle$ and $\langle\omega^2\rangle^{1/2}$ of the phonon spectrum of the alloys decrease as $x \to x_m$, i.e., a general softening of

the spectrum takes place, which can be explained only by intensification of the electron renormalization of the phonon frequencies in the case of an increase in the density of electron states ($\overline{E}$ increases with increase in x, see the Table). A similar phenomenon has been discussed in detail in Refs. 8 and 19.

A softening of the phonon spectrum near the limiting concentration of thallium has also been observed experimentally in Ref. 20, which is devoted to a study of the spectral functions of electron-phonon interaction in systems of In-Tl alloys. The reason for the increase in the electron parameter $\nu(\varepsilon_F)\langle I^2\rangle/M$ in both systems of alloys is apparently the Lifshitz transition, which takes place in In-Mg at $x_l \approx 0.02$ and in In-Tl at $x_l \approx 0.20$.

4. The intensification of the electron-phonon interaction leads to a crystal-lattice instability,[8] which is realized as a structural phase transition, and, since a temperature increase contributes to this process,[15] the boundary of the face-centered tetragonal phase on the solubility diagram should be shifted, upon decrease in the temperature, to the region of higher concentrations of the impurity, as is observed in practically all the indium alloys.[14] On the other hand, the increase in the electron-phonon interaction leads to a growth in the critical parameter of the superconducting alloys $In_{1-x}Mg_x$ and $In_{1-x}Tl_x$ at $x_l < x < x_m$.[3,10]

The fact that both the increase in T_c and the FCT $\rightleftarrows$ FCC transitions in systems with the divalen impurities Cd, Hg and Mg take place in nearby concentration regions, $x_m = 0.06$, 0.09 and 0.04, respectively, also attests to the essential role of the electron subsystem in the discussed structural phase transitions, in spite of the sharp difference in the ionic radii ($R_i^{Mg} \ll R_i^{Cd}$, R_i^{Hg}, Ref. 14). Upon introduction of the equal-valency thallium impurity, the Lifshitz transition (and the tetragonal distortion of c/a corresponding to it) sets in only at $x_l \approx 0.20$, while the structural phase transition occurs at $x_m = 0.23$.

4. CONCLUSION

The behavior of the lattice characteristics and of the parameters of the superconducting transition in $In_{1-x}Mg_x$ alloys is connected with the existence of the electron Lifshitz transition ($x_l \approx 0.02$), which precedes the first order structural phase transition. The experimentally observed softening of the shortwave region of the phonon spectrum is associated with the intensification of the electron renormalization of the phonon frequencies due to the increase in the electron parameter $\nu(\varepsilon_F)\langle I^2\rangle$ at $x_l < x < x_m$. The dependence of the critical temperature of the superconducting transition on the magnesium content in the face-centered tetragonal phase of the indium alloys with impurities of lower valence can be explained on the basis of these representations.

It can be assumed on the basis of analysis of the given experimental results that the phase transitions FCT $\rightleftarrows$ FCC in alloys of indium with lower-valence impurity in these systems are due to the intensification of the electron-phonon interaction, which is associated in turn with the behavior of the electron subsystem of these alloys.

In conclusion, we express our deep gratitude to A. A. Galkin for constant attention and support, and also V. G. Ba'yakhtar, N. B. Brandt, I. M. Vitebskiĭ, Yu. M. Kagan, E. G. Ponyatovskiĭ and B. Ya. Sukharevskiĭ for useful discussions of various problems encountered in this research.

[1)]It is known that upon introduction of an impurity of lower valence into the indium, a topological transition should take place initially, associated with the breaking of the β ring in the third Brillouin zone at the T points of the Wigner–Seitz cell, leading to a decrease in the density of states $\nu(\varepsilon_F)$ and a decrease in T_c.[10,11]

[2)]Data on $\omega_D(x)$ are lacking for the In-Mg alloys; however, this approximation corresponds to the experimental data for the other alloys of indium in the face-centered tetragonal (FCT) phase.[12]

[3)]A similar behavior of $g(\omega)$, but of both peaks, transverse and longitudinal, in the Pb-In alloy system, in which the frequency of the local oscillations lies in the range $\omega_0 > \omega_e$, was discussed in Ref. 18.

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