Thermopower of lithium–magnesium alloys at the $2\frac{1}{2}$-order transition

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The thermopower and resistivity of $Li_{1-x}Mg_x$ alloys in the concentration range $0 \leqslant x \leqslant 40$ at.% have been measured at various temperatures. At low temperatures a sharp thermopower peak is observed at $x_c = 0.2$ and is appreciably smoothed out at $T = 300$ K. The positive sign of the anomaly corresponds to the face of the Brillouin zone being touched by the Fermi surface. The anomaly in the resistivity is much less noticeable. The effect of the martensitic transformation on the results of the measurements is discussed. The ambiguity and spread of the results for specimens which do not undergo a martensitic transformation is noted. Hysteresis in the temperature dependence of the thermopower is observed near the thermopower maximum, the sign being the same as that for the body centered cubic phase.

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The work of Lifshitz[1] has appreciably stimulated the long-standing interest of physicists in studying the change in the topology of the Fermi surface which takes place when some controlling parameter such as pressure, anisotropic compression or impurity concentration, is changed. He showed that the thermodynamic properties and also the kinetic coefficients of a metal should have singularities of the type $|z|^{\pm 1/2}$ when there is a transition to a new topology (the formation or destruction of connecting necks, the appearance or disappearance of voids); z is here a parameter relative to which this transition takes place, and is proportional to the deviation of, for example, the compression or tension from the critical value where $z = 0$. According to the terminology introduced by Ehrenfest,[2] such transitions at a temperature $T = 0$ can be classified as $2\frac{1}{2}$-order phase transitions,[1] while for temperatures different from zero the transition is smeared and strictly speaking cannot be considered as a phase transition. Nevertheless, this nomenclature has recently become accepted in the literature to such phenomena.

A fairly large number of articles, both experimental and theoretical, have been published in recent years, devoted to studies of $2\frac{1}{2}$-order transitions. The details of the change in T_c of the superconducting transition under pressure was first observed by Brandt *et al.*[3] and served as the basis of the theory of Makarov and Bar'yakhtar,[4] who have shown that the $2\frac{1}{2}$-order transition must also manifest itself in a change in the superconducting properties of the metal. The majority of subsequent work[6–8] was concerned with measurements of the changes in the critical temperature of superconductors as a function of pressure and impurities (which, as has been shown,[5] can be considered as equivalent to some additional pressure). In general, the observations have been made on metals with a complicated Fermi surface (thallium, cadmium, indium), where there are transition points sufficiently close to the Fermi level to observe the transition in an accessible range of pressure and impurity concentration. Small irregularities (nonlinearities) are observed experimentally in the dependence of T_c on pressure impurities, and are associated with the expected $2\frac{1}{2}$-order transition. Watlington *et al.*[9] observed, in addition to the nonlinearity of T_c with the extension of filamentary cadmium crystals, some irregularity in the change of normal resistivity for the same values of deformation. Unfortunately, the anomalies which arise at T_c are certainly smeared out over a range of the order of the Debye temperature and are thus very weak in magnitude and difficult to measure.

In investigations of a somewhat different nature the main interest lay in studying the topological transition as such. For example, in the work of Brandt *et al.*[10,11] a bismuth single crystal underwent anisotropic deformations and, depending on the direction of the applied deformation, a transition was observed from three electron ellipses to two or to one, as fixed reliably by the Shubnikov–de Haas effect: the oscillation frequency of the corresponding section of one or other ellipse tended to zero as the deformation was applied; the crystal was not shattered and the picture was recovered reversibly on taking off the stress. However, no measurements were then carried out of physical properties in the behavior of which the anomalies accompanying the $2\frac{1}{2}$-order transition, and predicted by Lifshitz,[1] would be expected to appear. Mention should be made here of many observations of some or other properties of a metal or semimetal in topological transitions, which can be easily explained simply attributed merely to a change in topology, without invoking a hypothesis of radical anomalies: these are metal–dielectric transitions, changes in the components of the galvanomagnetic tensor by many orders of magnitude,[12] changes in magnetoresistance[13,14] when, for example, open trajectories arise, etc.

In relatively recent work on the stretching of filamentary crystals of bismuth[15] and aluminum,[16] in which record values of the relative deformations of a free crystal were achieved, at the same time as a definite cross section of the Fermi surface disappeared, indicating a $2\frac{1}{2}$-order phase transition, anomalies in the change of resistivity and also of T_c of aluminum were observed[16] for the same values of extension, and the anomaly in T_c was similar to that observed earlier in other work. It should be said that the value of the relative deformation of bismuth[15] at which one of the ellipsoids disappears agrees with that found in Refs. 10 and 11. In these, noted above, and in some other work, the anomalies ob-

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served in various T_c and resistivity dependences are extremely small and require some serious experimental problems to be overcome. On the other hand, the comparison of the observed anomalies with the basis features predicted by Lifshitz[1] also presents certain difficulties.

Attention was first paid in the recent work of Vaks *et al.*[17] to the fact that the anomalies which arise in a $2\frac{1}{2}$-order transition appear much more strongly in the thermopower than in other quantities. Characteristic anomalies are found in theoretical calculations of the behavior of resistivity and thermopower at a $2\frac{1}{2}$-order transition, produced by a change in electron concentration, as illustrated by Limg alloys; the anomaly in thermopower is relatively large.

The calculations in Ref. 17 were carried out on the basis of the pseudopotential method which describes both components Li and Mg.[18] However, no assumption were made about the possibility of the Fermi surface being smeared out (which, in general, gives the results[17] a sufficiently general character for them to be applied to experiments on $2\frac{1}{2}$-order transitions obtained by other means).

This circumstance increases appreciably the interest in the experimental observation of the predicted features at a $2\frac{1}{2}$-order transition, since the smearing of both the Fermi surface and the faces of the Brillouin zone proper, produced by nonuniformity in composition etc., makes the anomalies unobservable.[19] On the other hand, interest in just this system arises because $\mathrm{Li}_{1-x}\mathrm{Mg}_x$ alloys form a continuous series of solid disordered solutions with bcc structure over a great concentration range from $x = 0$ to $x = 0.7$ with relatively small changes in interatomic distances.[20,21] On the left of this sequence is lithium with a clearly closed Fermi surface which is somewhat different from spherical due to bulging in the direction of the twelve faces of the Brillouin zone. The Fermi surface of the free electrons already touches the zone boundaries for $x = 0.48$, so that in a real alloy the touching of the Fermi surface (without taking the smearing into account) should clearly occur and should take place much earlier. (According to calculations[17] in which the pseudopotential was constructed from a consideration of real data on the Fermi surfaces of lithium and magnesium, this value is $x_c = 0.247$.) Attention has therefore been paid to this system by experimentalists for a long time. From experimental results on positron annihilation,[22] Stewart in 1964 proposed that touching of the zone bondary by the Fermi surface occurs in the alloy $\mathrm{Li}_{81}\mathrm{Mg}_{19}$. Measurements were later made of the Hall voltage,[23] optical properties,[24] and Knight shift.[25] From various considerations (for example, a small smooth irregularity in the change of Hall constant) a value $x_c \gtrsim 0.3$ was assigned. The conclusions in these works were drawn by using very involved interpretations of the experimental facts and are thus not very reliable. The same value $x_c = 0.19$ as in Ref. 22 was obtained from magnetic susceptibility results.[26] As far as we know, the resistivity and thermopower of LiMg alloys have not been measured before, and the question of the observation of square-root singularities[1,17] remains open.

All work on LiMg alloys was carried out at liquid-nitrogen temperatures or above. This is because in these alloys, as in pure lithium, a martensitic transformation takes place which never proceeds to completion. There is thus a mixture of two phases below the transition temperature M_s: bcc and hcp. In pure lithium $M_s = 70$ K. By carrying out the experiment at temperatures $T \geqslant 78$ K, only the bcc phase could thus be studied. In fact, however, M_s rises fast on increasing x;[27] for example, for $x = 0.12$ we have $M_s = 125$ K.[28] Thus even at liquid-nitrogen temperatures for $x \geqslant 0.02$, i.e., for practically all concentrations, there is a mixture of two phases and for concentrations $x > 0.1$, i.e., those of most interest, the ratio of the volumes of the bcc and hcp phases are practically unchanged for $T < 78$ K. Thanks to this it was possible to carry out measurements both at nitrogen and helium temperatures where there is no thermal smearing of the properties look for. In addition, at low temperature the electronic part of the thermopower can be separated out sufficiently reliably. Preliminary resutls of the experiments have been published earlier.[29]

THE EXPERIMENTS

The LiMg specimens were melted together in a pure argon atmosphere in an induction furnace without using a crucible.[30] A molten drop, of spherical shape, was released into a cold demountable copper mold where the alloy rapidly solidified. The magnesium concentration x in the ingot was determined from the weight in the initial batches. In addition, part of the ingot was analyzed chemically. The 0.6 mm diameter wire measurement specimens, 30–40 mm long, were obtained by extrusion through a die. Several control specimens were sometimes prepared from one ingot. We must point out the good agreement between experimental results for different specimens from a single ingot, and also the agreement between the results of chemical analysis and the concentration determined by batch weighing. The disagreement in the magnitude of the concentration did not exceed ± 1.5 at.%.[29] This error, which is much greater than the accuracy of the batch weighing, is evidently due to the different melting regimes (time, temperature) and to the components evaporating at different rates.

Lithium and its alloys with magnesium, especially for small x, are very active chemically. All procedures, therefore, were carried out on specimens in an atmosphere of argon (in a sealed box) and helium. Pure indium solder was used for the electrical and thermal contacts. Beryllium bronze spring contacts were also used for resistance measurements. The impurity content in the initial components, lithium, and magnesium, was not more than 0.005 at.%. The resistivity ratios were $\rho_{300}/\rho_{4.2} = 700$ for the initial lithium and ~ 300 for magnesium. On melting in the oven, some uncontrollable amount of impurity must inevitably fall into the alloy, for example from the surface of the billet used. This produces a perceptible effect on the final result of the measurement of both resistivity and thermopower. The resistivity ratio for the control alloy $\mathrm{Li}_1\mathrm{Mg}_0$, i.e., for pure lithium which had undergone all the procedures for preparing the alloy specimens, but without the addition of magnesium, was $\rho_{300}/\rho_{4.2} = 400$. The magnitude of the thermopower for this specimen also differs noticeably from that for the pure initial lithium. Nevertheless, this addition is much less than the magnitude of the effect observed. Besides, the procedure was identical for preparing all specimens, so that it can be supposed that the uncontrolled impurities could not affect the results obtained appreciably.

The thermopower was measured by a differential method relative to pure lead, for which the values are known.[31] The LiMg specimens, the lead wire and also a copper–constantan thermocouple were soldered to copper foils which were stuck with BF-2 adhesive to two copper plates between which a temperature difference $\Delta T \approx 0.5$ K was established. Both these plates were inside a copper capsule with a common heater and there was also a thermometer inside. The whole apparatus was housed in a sealed finger, filled with helium heat-exchange gas at a pressure chosen to fit the conditions of the experiment. The temperature could thus be controlled over a fairly wide range. The temperature drop was provided by two identical heaters stuck to the copper plates mentioned above, which were switched on in turn so that the power dissipated in the capsule remained constant. The direction of the heat flow in the specimens and thermocouple was thus reversed (as is usually done when measuring small resistances). The temperature differences ΔT obtained in this way are of opposite sign, but slightly different in magnitude due to some unavoidable asymmetry in the assembly and external temperature gradient (in the cryostat). Nevertheless, the uncontrolled zero drift was eliminated in this way and the limiting sensitivity of ≈ 0.5 divisions could be used (a R363-3 potentiometer with sensitivity $\approx 10^{-8}$ V/div was used). This sensitivity could, in fact, only be achieved at helium temperatures, where because of the small heat capacity of the system the time for establishing thermal equilibrium on reversing the heat flow is less than the characteristic measuring time. The sensitivity is worse at high temperatures (but th signal is much larger), and the errors in measurements are also noticeably increased at temperatures above T_c for lead, where the thermopower of lead has to be taken into account. The sign of the measured thermopower was monitored by the jump in thermopower of lead at the transition to the normal state.

RESULTS OF THE MEASUREMENTS

Measurements of thermopower and resistivity were carried out on more than twenty specimens of $\mathrm{Li}_{1-x}\mathrm{Mg}_x$ alloys in the concentration range $0 \leqslant x \leqslant 40$, where x is the Mg content in at.%. The temperature dependences of the thermopower α for several specimens are shown in Fig. 1, where the points for different specimens are indicated by different symbols. The corresponding magnesium concentration x is shown to the right of each set of points. The points for $x' = 0$ and $x = 0$ correspond to the pure initial lithium (open circles) and lithium which went through the remelting procedure (triangles).

The striking difference in slope of the plots obtained show the strong concentration dependence of the electronic part of the thermopower. It should, however, be pointed out that all the curves do not go exactly to zero as they should, and we ascribe this to some systematic error. The concentration dependence of the mean slope at helium temperatures is shown by curve 1 (full circles) in Fig. 2. Following the arguments discussed above, the value of the thermopower of the control "alloy" $\mathrm{Li}_1\mathrm{Mg}_0$ was used for $x = 0$. The results of measurements of thermopower at $T = 78$ K are shown as curve 2 (triangles) in the same figure. The value of α/T is the ordinate so that if the $\alpha(T)$ dependences were linear, curves 1 and 2 should fall on top of one another. The appreciable difference in the curves is evidently produced by a phonon-drag contribution varying monotonically with concentration. It is difficult to compare quantitatively the magnitude of the thermopower peak for $x_c = 0.2$ on curves 1 and 2, since the position of the "zero" on curve 2, i.e., the magnitude of the phonon contribution, is difficult to determine.

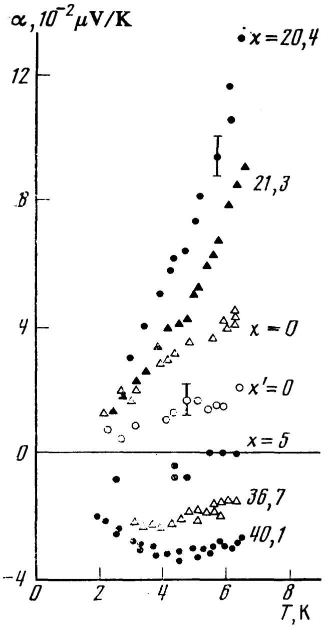

FIG. 1. Temperature dependence of thermopower for various specimens with magnesium concentration in at.% shown to the right of each plot.

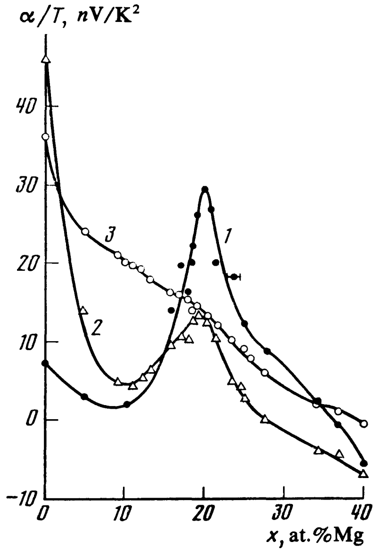

FIG. 2. Dependence of thermopower on magnesium concentration for various temperatures: 1 (●)—$T \sim 4.2$ K; 2 (△)—$T = 78$ K; 3 (○)—$T = 300$ K.

Nevertheless, it can be concluded that these quantities are of the same order of magnitude. At first glance it might appear that the maximum of curve 2 is shifted in the direction of smaller values of x. However, considering the monotonic fall in the phonon contribution with concentration, it can also be considered that $x_c = 0.2$ at $T = 78$ K. The phonon contribution evidently becomes dominant at room temperature and the anomaly is hardly visible—curve 3 (open circles).

The resistivity was also measured for all specimens at various temperatures. The ratio $\rho_{300}/\rho_{4.2} \approx 2$ for concentrations $x \sim 0.2$ and the value $\rho_{4.2} \approx 2 \times 10^{-5}$ Ω·cm. No anomaly in the $\rho_T(x)$ variation could be found. Unfortunately, the absolute error in measurements of both ρ_{300} and $\rho_{4.2}$ was $\sim 5\%$, mainly due to the imperfection of the specimen geometry and to the error in determining the distance between the potential contacts. Geometrical factors are eliminated in the $\rho_{300}/\rho_{4.2}(x)$ variation and the relative error is not more than 1.5%. At $x = 0.2$ some irregularity is then noticeable which is outside the limits of experimental error (curve 1 of our earlier note[29]), but this curve is the results of dividing two variations, $R_{4.2}(x)$ and $R_{30}(x)$, each of which has apparently a singularity; it is therefore difficult to interpret their ratio. It must just be pointed out that the resistivity results illustrate well the fact that the $2\frac{1}{2}$-order singularity appears much more strongly in the thermopower than in other properties.

It sould be stated that all the initial thermopower measurements were first carried out in liquid helium, and then the same specimens were measured at nitrogen and room temperatures. Our earlier results[29] and also the results shown in Fig. 2 were obtained on making the measurements in the same sequence. A completely different picture is observed if the thermopower is first measured at room and nitrogen temperatures before the specimen is cooled to helium temperatures and these results are compared with those obtained on these same specimens after a cooling cycle. The disagreement is most marked near the transition point $x = 0.2$ at room temperature and to the left of this point at nitrogen temperature. This is illustrated in Fig. 3, where the open circles and triangles correspond to specimens never cooled to helium temperature, while the full circles and triangles are the results of measurements on the same specimens which were cooled just once to helium temperature, i.e., corresponding to curves 3 and 2 in Fig. 2. As can be seen from Fig. 3, the scatter of points after cooling is appreciably less. It is also interesting to note that if the concentration dependence of the thermopower at nitrogen temperatures is plotted for the original uncooled specimens, the maximum is shifted considerably to the left, to $x_c' \sim 0.17$. Such a difference and the multivalued results are most likely to be explained by the freshly prepared specimens having some differences in the inhomogeneity structure, which furthermore influence the nature of the course of the martensitic transition. The thermal cycling to helium temperatures, however, leads to equalization of the inhomogeneities (similarly to annealing).

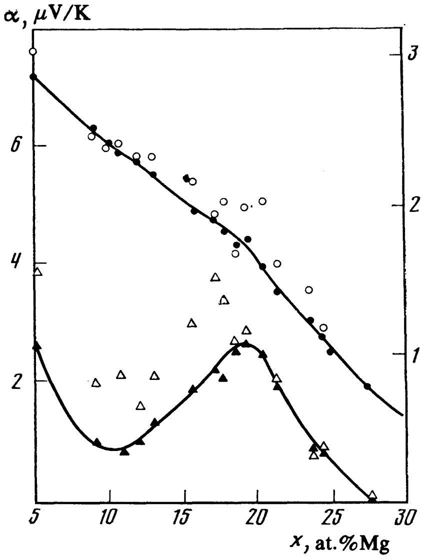

FIG. 3. Results of measurements of thermopower of specimens not previously cooled to helium temperatures—open circles ($T = 300$ K, left hand scale) and open triangles ($T = 78$ K, right hand scale)—compared with measurements of thermopower of the same specimens after cooling—full circles and triangles, i.e., corresponding to curves 3 and 2 of Fig. 2 but plotted differently.

As has already been mentioned, LiMg alloys at nitrogen and lower temperatures consist of two phases: bcc and hcp. Although the observed thermopower anomaly occurs up to room temperature where there is no hcp phase, and this could the taken as indirect confirmation that the observed anomaly is produced by the bcc phase, it would, nevertheless, be interesting to try to confirm this fact more directly. The reverse transition from hcp to bcc on raising the temperature takes place later and leads, as already mentioned above, to a phase-ratio hysteresis which is specially large at small magnesium concentrations. Since the relative content of the bcc phase is larger in the cooling part of the thermal cycle of the hysteresis and smaller in the heating part, this quantity traces an anticlockwise hysteresis loop. The relative content of the hcp phase for the same reasons traces clockwise hysteresis. It is natural to presume that if the thermopower of one or the other phase is mainly responsible for the anomaly in the electronic component, then the measured thermopower will also undergo hysteresis on thermal cycling, with the sign of the hysteresis the same as that of its own phase. It is essential that the change in the phonon component should then be rather small. The concentration $x \approx 0.17$ was chosen for the measurements since it could be expected from the results of the measurements (Fig. 2) that the phonon contribution is a minimum in this concentration region (since the value of α/T is of the same order of magnitude at all temperatures).

Measurements of thermopower in thermal cycling are shown in Fig. 4. The upper plots correspond to measurements on two specimens (circles and triangles) from one billet with $x \approx 0.17$. The full symbols correspond to heating and the open symbols to cooling. In addition, the direction of the path is indicated by arrows. The sign of the observed hysteresis is anticlockwise and is evidence that the thermopower in the bcc phase is larger and that, consequently, the $2\frac{1}{2}$-order transition is observed in this phase. Similar thermopower measurements on thermal cycling of a specimen with

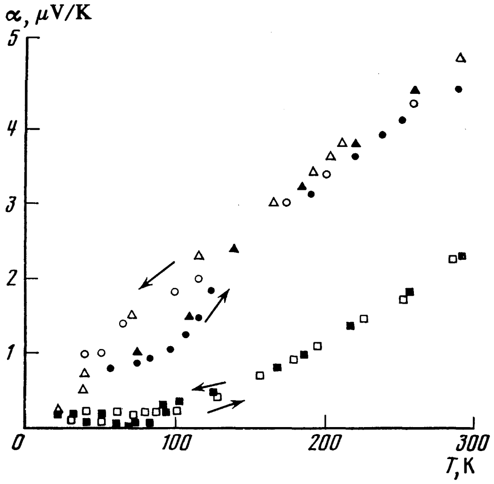

FIG. 4. Temperature dependence of thermopower on heating (full symbols) and cooling (open symbols). Circles and triangles (above)—two specimens from a billet with $x = 0.17$; squares (below)—$x = 0.24$.

$x \approx 0.24$ are given lower down for comparison; the value of α is the same at helium temperatures as for the previous specimens. It can be easily seen that the phonon contribution is high and negative and hysteresis is not observed.

As already mentioned, the martensitic transition in LiMg alloys occurs in the same way as in lithium, but the existing information[27,28] pertains to small magnesium concentrations, i.e., to the left of the $x_c = 0.2$ transition. There is some indication that the hysteresis decreases with increasing x. It would thus be of general interest to trace the nature of the change in the martensitic transformation for $x \gtrsim 0.2$. For this purpose an alloy at the maximum thermopower was first studied. One of the plots of a measurement of the acoustic emission[32] which occurs in the martensitic transition for slow cooling[1)] is shown in Fig. 5 for a specimen with $x \approx 0.20$. In contrast to pure lithium, for which a large peak in the acoustic emission is observed[32] at the martensitic transition, and the whole transition occurs over an interval $\Delta T \sim 10$ K, in this alloy the transition is accompanied by a large number of weak peaks spread over a temperature interval from 210 to 130 K. Such a behavior points to an extremely weak rate of change of the structure.

On the other hand, therefore, the martensitic transition becomes much smoother and drawn out over a range of $T \sim 100$ K, while on the other the magnitude of the hysteresis, i.e., the difference in the ratio of phases on the outward and return paths, does apparently actually decrease appreciably. In other words, the martensitic transformation becomes more reversible. For this reason hysteresis is not observed in measurements of thermopower for a specimen with $x = 0.24$ (Fig. 4).

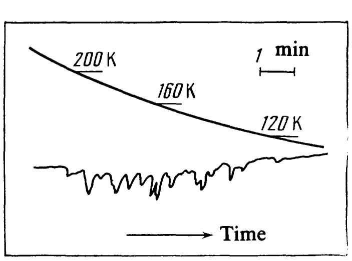

FIG. 5. Trace of the time variation of acoustic emission: lower trace—on coolling a $Li_{80}Mg_{20}$ alloy, upper trace—temperature variation.

CONCLUSIONS

An anomaly was observed in the concentration dependence $\alpha(x)$ at $x_c = 0.20$, which can be traced up to room temperature where this anomaly is very smeared out. From the nature of the observed $\alpha(x)$ dependence at low temperatures it follows that the smearing of the Fermi surface or the boundaries of the Brillouin zone is disordered alloys can be sufficiently small for an electronic $2\frac{1}{2}$-order transition to be very clearly seen. The positive sign of the peak in α $(x = 0.2)$ corresponds to the Fermi surface touching the Brilloiun zone boundary at this concentration.

By comparing the experimental results with the results of calculation,[17] it can be established that the critical concentration $(x_c^{\text{th}} = 0.247)$, and the form of the $\alpha(x)$ dependence to the left of the maximum are in good agreement, also that the anomaly in thermopower α is much larger than that in the resistivity. The disagreement in the shape of the $\alpha(x)$ plot on the right of the maximum shuld be noticed: instead of the sharp fall obtained theoretically, a fall with about the same curvature as the growth to the left of the maximum is observed. Two possible reasons for such a behavior can be proposed. First, real inhomogeneities an the smearing of the Fermi surface should lead, due to strong scattering, to a more symmetrical and finite peak in thermopower compared with the "ideal" shape of the theory.[17] Second, as a neck appears, a group of electrons arises with dynamics of motion described now not by a convex Fermi surface but by a concave-convex surface, i.e., the dynamics acquires a partly "hole" character. The formation of a pure "hole" band should lead to the anomaly in thermopower such as is shown on the inset to Fig. 2, b of Ref. 17, but of opposite sign, i.e., to a positive peak with a gentle descent on the right of the transition, which is observed.

The anomaly is observed at all temperatures, so that it is natural to assume that it is associated with the bcc phase, since there is no hcp phase at room temperature. Measurement of the thermopower hysteresis, the sign of which coincides with the sign of the change in the bcc phase, is also evidence that the observed peak in α corresponds to the bcc phase. There should also be a topological transition in the hcp phase as x is increased. From the free electron theory, touching of the Fermi sphere by the side faces of the Brillouin zone occurs for $x = 0.14$, and by the end faces (in the [0001] direction) for $x = 0.36$ (faces of the 2nd Brillouin zone, i.e., faces of the Jones cell). It can, therefore, be assumed that for the real Fermi surface of the hcp phase there is already touching of the side at $x = 0$. It should be noted that touching of the end faces should occur within the measured concentration range but is not observes separately.

If it is assumed that the concentration singularity observed in the thermopower somehow corresponds to a singularity in the density of states, then the size of this singualrity, in terms of energy, turns out to be somewhat less then (roughly half) the magnitude of the gap, so it is natural that at $T = 300$ K the singularity should be smeared out. However, it is observed quite sharply at low temperatures, although the conductivity is only doubled. This can be explained by the diffuseness of the Fermi energy being $\Delta\varepsilon = \hbar/\tau_E$, where τ_E is the energy relaxation time, which is not less than the momentum relaxation time τ_{tr}. At high temperatures $\tau_E \sim \tau_{tr}$. At low temperatures $\tau_E \gg \tau_{tr}$ and the diffuseness becomes appreciably less.

The observed ambiguity and the scatter of the points in measurements of freshly prepared specimens, which become completely smoothed-out after cooling at helium temperatures indicates the existence of a considerable effect of prior history (the temperature regime) on the results of the measurements. This observation, which is undoubtedly interesting in itself, means that one must exercise a certain caution in comparing experimental results of different investigations.

In conclusion, we wish to draw attention to the strong concentration dependence of the phonon contribution to the thermopower, which decreases as x is increased and appears to change sign. The thermopower anomaly is smoothed out at $T = 300$ K and a monotonic component dominates the $\alpha(x)$ dependent. This behavior can in general be used for an accurate determination of magnesium concentration by measuring the thermopower of specimens previously cooled to helium temperatures, using curve 2 of Fig. 2 as the calibration.

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