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INFLUENCE OF THE PLASTIC DEFORMATION ON THE CHARGE CARRIERS CONVERSION IN HIGH ENTROPY ALLOY $Al_{0.5}$CoCuCrNiFe

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In range ~300…77 K the anomalies of the fundamental transfer coefficients of high-entropy $Al_{0.5}$CoCuCrNiFe alloy, as well as influence of plastic deformation on these anomalies were studied. Both the anomalies themselves and the deformation effects are associated, we believed, with hole↔electron conversion, which changes the ratio of the areas of electron and hole of the Fermi surface sheets. The kinetic data are compared with the temperature behavior of the spontaneous magnetization (I_s) of the equiatomic AlCoCuCrNiFe alloy, which contain a set of 3-d ferromagnets in almost the same proportions. Interest in AlCoCuCrNiFe is caused by presence of the anomaly in I_s(T) in the same temperature range where the kinetic features of $Al_{0.5}$CoCuCrNiFe we observed. The explanation this magnet anomaly was given by the phenomenon of hole↔electron conversion under the assumptions, that i) I_s has electronic (I_{se}) and hole (I_{sh}) components, and ii) the quanta of these components (i_{se} and i_{sh}) satisfied the condition $i_{se}>i_{sh}$.

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INTRODUCTION

Until now, the interest in the high-entropy alloy (HEA) $Al_{0.5}$CoCuCrNiFe was mainly due to possibility of its using at cryogenic temperatures (T) owing the unique combination of high strength and ductility [1, 2]. Limitation of such point of view was demonstrated in [3], where the question was raised about of previously untouched scientific aspect. The fact is, that along with the nonlinear increasing in strength below T~220 K [1, 2], (Fig. 1,a), the anomalies of the kinetic and acoustic coefficients were observed [4, 5]. The nature of the latters was explained [3] by the phenomenon of hole↔electron conversion (HEC) in a two-component system of charge carriers. In addition to possibility of discovering a connection between strength properties and HEC, interest in such research is associated with presence of the ferromagnetic transformation in the alloy below T~350 K with dependence of spontaneous magnetization (I_s) on T [6] that contradicts theoretical concepts.

The objective of this work was to substantiate the HEC hypothesis put in [3] for system of two types of charge curriers: negative charged electrons (electron-like, e) and positive charged holes (hole-like, h). For this purpose, the differences in the temperature behavior of the same as in [3] transfer coefficients for samples of $Al_{0.5}$CoCuCrNiFe (absolute thermopower, S, electrical conductivity, Ω, thermal conductivity, k) caused by plastic deformations (ε) were studied. The choice of ε as a influencing factor on above coefficients – first of all on S, – is due to the results of works [7, 8], in which the dependences S(T) of annealed and deformed copper was studied. These works shown that sign of S of copper in both states changes with decreasing T in vicinity of ~100 K from "+" to "–". That indicates the loss of the prevalence of negative charge carriers was existed above ~100 K, i.e. on charge carriers' conversion of type e→h. The second important result of [7, 8] is discovery of the effect of a change in the sign of S_{Cu} in the conversion range as a result of plastic deformation.

We hoped registration of the deformation effect in the temperature behavior of kinetic coefficients of $Al_{0.5}$CoCuCrNiFe HEA would become an argument in favor of validity of the concept being developed. In this case, it is advisable to connect the anomalies in the mechanical and dissipative properties of $Al_{0.5}$CoCuCrNiFe [1, 5] with the concentration of charge carriers of the electronic and hole types, which, according to [9], determines the type of basic crystal structures (fcc and bcc) in HEAs., i.e. is a factor influencing the mechanical properties of metal [10, 11].

Of particular interest was the correlation between the kinetic anomalies of this alloy and the temperature change anomaly in I_s of the HEA composition AlCoCuCrNiFe we visibled.

SAMPLES AND METHODS

Samples of $Al_{0.5}$CoCuCrNiFe HEA, obtained by fusing components (at.%: Al – 4.46; Co – 19.48; Cu – 21.01; Cr – 17.18; Ni – 19.4; Fe – 18.46) in an argon atmosphere [12], were cut by electric spark cutting plates ~50×3×0.7 mm. To homogenize their state, hereinafter called the initial state, they were annealed for 12 hours at T≈1250 K. Measurements of above temperature dependences in the range of ~300…77 K were carried out on samples in four states, including the initial one. The second, third and fourth states are obtained by successively increasing the degree of plastic deformation by rolling ε=9, 43, and 58.5% along the long axis of the sample at T~300 K.

Temperature T was measured by a Cu-Const thermocouple with an accuracy of ~0.5 K. Conductivity Ω was calculated from the electrical resistance value R=1/Ω measured using a 4-point circuit (accuracy no worse than 1%). The value of absolute thermopower S was taken to be S=δU/δT, where $\delta U=U_1–U_2$ is the potential difference (measurement accuracy $\sim 3\cdot 10^{-8}$ V), established between the sample ends at a temperature difference $\delta T=(T_1-T_2)\sim(5...10)$ K. The appearance of δT was a consequence of heating one of the ends by a heat flux from a miniature electric heater powered by a direct current I=const. The sensitivity of δT measurements, carried out by a pair of oppositely connected Cu-Const thermocouples, was $\sim10^{-3}$ K.

Image on the character of the k(T) dependence was obtained using developed by us [13–15] version of the classical method of uniaxial stationary heat flow [16], which does not require taking into account heat losses. In this technique, the adequacy of the results is ensured by measuring the dependence $\delta T(T)$ under conditions of a strictly monotonic change in all heat losses in changing T. Strict compliance with the last condition (see [13–15]) allows us to consider the relation $\delta T(T) \propto 1/[k(T)\times\Psi(T)]$, where $\Psi(T)$ is monotonic function, to be valid. Denoting $[k(T)\times\Psi(T)]\equiv k^*(T)$, where $k^*(T)$ has the meaning of a monotonically scaled real thermal conductivity coefficient k, we obtain a representation of the qualitative dependence of k(T) using the $k^*(T)$ curve.

Numerated dependences were measured in the mode of a stepwise decrease in the temperature of the sample, which was in a vertically oriented Dewar vessel in nitrogen gas vapor. The configuration of the isotherms of such a thermal field ensures (see [13–15]) the monotonicity of the decrease in T of the sample as it approaches the liquid level monotonically and, at the same time, the monotonicity of the coefficient $\Psi(T)$. As a result of a large number of measurements, we came to the conclusion – this is important to emphasize – that to obtain adequate results, two steps of thermalization procedures ~ 10 min are required, including thermalization of the initial state (Q=0, δT=0) and thermalization after each heat flow supply.

RESULTS

Fig. 1 shows the graphs of the normalized dependences $\Omega(T)/\Omega_0$, $S(T)/S_0$, and $(k^*(T)/k^*_0)$ (T_0=295 K, ε=0; 9; 43; 58.5%), belonging to the same sample. When describing and analyzing the anomalies observed approximately in the middle of the studied temperature interval ~300…77 K, we agree to consider the case of a decrease in T. Note also that in the indicated temperature interval all samples have S>0, which indicates the prevalence of electron-like curriers, [17], i.e. $(N_e/N_h)>1$.

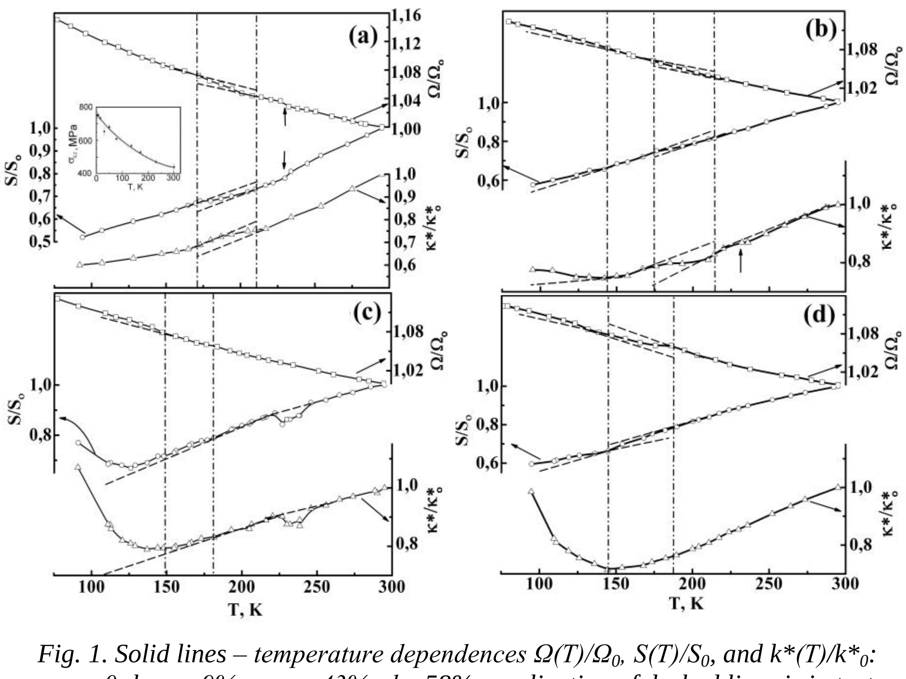

Fig. 1. Solid lines – temperature dependences $\Omega(T)/\Omega_0$, $S(T)/S_0$, and $k^*(T)/k^*_0$: a – ε=0; b – ε=9%; c – ε=43%; d – 58%; application of dashed lines is in text. Insert in Fig. 1,a – temperature dependence of the conditional yield strength $\sigma_{0.2}$ [1]

Let's start with an analysis of the features of the curves in Fig.1,a, where the dashed lines show linear extrapolations of "non-anomalous" sections of curves to the anomaly interval. Thus we reveal a range of ~210…170 K, within which the values of the derivatives $dS(k^*, \Omega)/dT$ differ noticeably from the values both above T~210 K and below T~170 K (we consider these values to be the entry into the anomaly and the exit from it). As for the signs of anomalies, i.e. signs of the differences between the experimental and extrapolated values, for example, $S(T)^{exp}-S(T)^{extr}$, then the input anomalies S and k* have a "+" sign, and Ω – "–". This means that the consequence of entering in anomalous interval from higher temperatures is a lowering in the rates of decrease of S and k*, as well as a rising in the rate of increase in conductivity Ω. At T~170 K, all three curves also exhibit features. Their nature makes it possible to interpret the processes in the crystal structure as reduction of anomalies, i.e. partially returning derivatives to pre-anomalous values. Concluding the description of Fig. 1,a, we note that two of its curves have weak features in the vicinity of ~230 K. They will be discussed separately.

The graphs in Fig. 1,b, ε=9%, have both similarities and differences of the characteristics compared to the initial state (see Fig. 1,a). Thus, each of the curves in

Fig. 1,b has an input anomaly at T~210 K with the same sign as above. However, certain features emerged. For example, after an approximately linear course below ~170 K, all curves begin to gradually "lift up" from T~145 K.

The curves in Fig. 1,c, ε=43%, do not have input anomalies at T~210 K, but at T~180 K the input anomalies appeared, the sign of which is similar to the sign of the anomalies of the curves for ε=0. This looks like a deformation shift of the input anomaly of the "same sign" in the temperature in the direction of low T. We also note that the nature of the curves noticeably changed below T~145 K, indicating an increase in the influence of deformation on the kinetic coefficients. As for the features in the vicinity of T~230 K, they are in the form of "returning" anomalies in $S(T)/S_0$ and $k^*(T)/k^*_0$ in a temperature range of T~15 K.

The most radical change in the manner of the curves is demonstrated in Fig. 1,d, relating to the structure formed by deformation ε=58%: input anomalies are disappeared at T~210 K, "returning" anomalies in the vicinity of T~230 K are disappeared too. At the same time, in-phase anomalies with sign " −" are appeared in the interval T~180…145 K. At the end of this interval (T~145 K), the signs of all anomalies change from "–" to "+" and do not change until T~77 K. The intensity of the anomalies, as can be seen, increases below T~145 K. As for quantitative changes, deformation leads to an increase in S_0 from ≈(3±~0.3) μV/K (ε=0) to ≈(6.0±~0.3) μV/K (ε=58%).

Presented data allow to assert the presence of deformation effects (DE) in anomalies in the conductive, thermoelectric and heat-conducting properties of $Al_{0.5}CoCuCrNiFe$ below T~210 K. These effects consist of a shift towards low temperatures in the temperature range of conversion anomalies with increasing ε and a twofold change in the direction of conversion processes.

DISCUSSION

The terms "hole" and "hole-like conductivity" owe to possibility of formally to describe thermo- and magnetoelectric phenomena using the assumption about of presence of both negatively and positively charged quasiparticles among current carriers.The latter, hole-like, h [9, 18, 19], have a positive charge equal to the electron charge and a certain effective mass. In quantum theory their value is determined by the local curvature of the valence band [18]. It is believed that the positivity of the hole charge is a consequence of the presence of the electron itself in the crystal lattice under the complex influence of atomic forces. Proof that in reality charge transfer in all cases is carried out by electron-like carriers was obtained in measurements of the gyromagnetic effect. In our discussions, however, we will almost always use a more convenient phenomenological description.

Let us recall several useful provisions for further understanding:

1. The absolute thermopower coefficient S, which characterizes the physical properties of a substance, is defined as the ratio of the potential difference between the ends of the conductor to the difference in their temperatures: $S=\delta U/\delta T$.

2. It is believed that electrons and holes make independent contributions to σ and S [18]:

$$\sigma=n_e|e|u_e+n_h|e|u_h, \qquad (1)$$

$$S=S_e-|S_h|. \qquad (2)$$

In (1), the symbols $n=N/v_0$, where v_0 is the specific volume, and u with the corresponding indices denote the densities and mobilities of electrons and holes.

3. For electronic conductivity, S>0 and in the case of hole conductivity, S<0 [17]. From (2) it is clear that sign of S is determined by the prevailing carriers.

4. The value of N_e/N_h, which determines the sign of S, depends on T [17], up to a change to the opposite, what is interpreted as a transition of the prevalence from one type of carrier to another due to the phenomenon of conversion [17–19].

5. In addition to the case of obvious conversion, when the sign of S changes to the opposite, a situation is possible when S(T) exhibits an anomaly that does not change the sign of S. One of the reasons for this may be the so-called latent conversion (LC), as a result of which the ratio between N_e and N_h changes. Another reason for the anomaly with sign S=const is the ST, which changes the effective scattering cross section of carriers. The reasons are easily distinguishable: the consequence of ST should be a correlating anomaly in the dependence Ω(T) of the opposite sign than the sign of the anomaly S(T).

When analyzing the results, it was taken into account next.

I. $Al_{0.5}CoCuCrNiFe$ samples in the studied states own typically metallic dependences Ω(T) and S(T) with (dΩ/dT)<0 and (dS/dT)>0 (see Fig. 1).

II. There are no STs below ~228 K in annealed $Al_{0.5}CoCuCrNiFe$ samples, as evidenced by the monotonicity of Young's modulus [2].

III. In HEAs of the AlCoCuCrNiFe family, the changes in the sign of S were observed with changing T [20], on the basis of which we can assume $S=(S_e-|S_h|)>0$, $N_e/N_h>1$.

Initial state, Fig. 1,a. Based on II, the interpretation of the anomaly in the range of ~210…170 K as caused by a change in the effective scattering cross section of carriers due to ST is excluded. Under the alternative explanation (i.e., LC), two experimental findings indicate the types of input and output conversions. In fact: i) a decrease in the derivative $[dS(T)/S_0]/dT$ without of changing the sign of S at the entrance to the anomaly and its increase at the exit; and ii) the approximate linearity of the anomalous part of curve indicates h→e and e→h types of LC, respectively.

Let us analyze the response of the coefficients Ω and k* to conversion. We remember that the effective scattering cross section, even in the case of the same crystal structure, is different for carriers having different of u [19]. We see (see Fig. 1,a) that at the entrance to the anomaly the growth rate of Ω increases, but at the exit it decreases. Since at the input, as a result of the h→e conversion, the N_e/N_h ratio increases, it follows that the input anomaly is associated with an increase, and the output anomaly, with a decrease in the number of electron-like carriers in the ensemble of charge carriers. It follows that the HEC concept is consistent with experiment if $u_e > u_h$. The relationship between u_e and u_h in our case is unknown. Confidence in the correct interpretation of the essence of anomalies based on the LC concept is given by the sign of the input anomaly of the $1/\Omega$ dependence, Fig. 1,a – it excludes a ST.

Let us write $k \equiv k^*/\Psi$ (see above) in the form

$$k = k_e + k_h + k_{ph}, \quad (3)$$

where the symbols k_e, k_h, and k_{ph} denote the charge (e and h) and phonon (ph) components of the total k. We see in Fig. 1,a that at the entrance to the anomaly the temperature decline in k slows down, but at the exit the picture is the opposite. If $u_e > u_h$, then the input effect can naturally be attributed to the total charge component $k_e + k_{kh}$, which has become more mobile, while at the output the mobility should decrease slightly.

Deformed samples. The curves in Fig. 1,b are similar to the curves of the Fig. 1,a, therefore we interpret their anomalies in the range of ~210…170 K within the framework of the same concept of input and output LCs of opposite directions. We see that the agreement between the signs of the anomalies of the coefficients Ω and k* and the relation $u_e > u_h$, necessary for the realism of the concept, is saved.

As a result of deformation by ε=43% (see Fig. 1,c): i) the temperature of the input anomalies decreased to ~180 K, ii) the cause of the input anomalies is still the h→e conversion. Now, however, the conversion of the same character continues with decreasing T, intensifying below ~145 K. As in Fig. 1,a,b, the signs of the anomalies of the curves $\Omega(T)/\Omega_0$ and $k^*(T)/k^*_0$ correlate with the inequality $u_e > u_h$.

Fig. 1,d illustrates the dramatic changes in the curves. Thus, the dependence $S(T)/S_0$ does not have a sign-positive anomaly in the range of ~180…145 K; in its place a sign-negative anomaly appears, which turns into a sign-positive anomaly at T~145 K. Within the framework of the LC concept, we conclude that at the entrance to the sign-negative anomaly a conversion of the form e→h occurs, and at the exit, coinciding with the entrance to the low-temperature sign-positive anomaly, the type of conversion changes to the opposite, i.e. to h→e. In this case, the signs of the input and output anomalies of the dependences $\Omega(T)/\Omega_0$ and $k^*(T)/k^*_0$ still correspond to the relation $u_e > u_h$. In other words, a change in the direction of conversion leads to an inversion of the signs of the anomalies of the coefficients Ω and k. We consider this fact to be a decisive argument in favor of the validity of the LC hypothesis.

Anomalies in the vicinity of T~230 K. According to data [1, 2], the interdendritic spaces of annealed $Al_{0.5}$CoCuCrNiFe alloy contain particles of harder phases that precipitated during annealing. They initiate appearance of the Koiwa-Hashiguchi peak in the temperature dependence of internal friction at T~230 K and a feature in the temperature dependence of the Young's modulus. In addition, annealed $Al_{0.5}$CoCuCrNiFe has in the vicinity of T~230 K the anomalies of the opposite sign in the $\Omega(T)/\Omega_0$ and $S(T)/S_0$ dependences [4]. Therefore, the anomalies at T~230 K cannot be attributed to the LC of charge carriers and therefore were not the subject of study in this work.

Thus, as a result of the analysis, it was established:

1. The deformation effect in the temperature behavior of the kinetic coefficients S, Ω, and k of the $Al_{0.5}$CoCuCrNiFe HEA is similar to the same effect in [7, 8]. This is evidenced by i) a shift in the temperature range of the anomaly as a result of plastic deformation by rolling and ii) a change in its sign at a sufficient value of ε.

2. The signs of the anomalies of the coefficients S and Ω at the ends of the temperature interval of the anomalies change to the opposite when the type of anomaly is inverted.

Both conclusions allow to consider the concept of LC proven.

3. From the established factors influencing conversion processes (temperature, deformation) in HEAs, it follows that the fundamental cause of the conversion phenomenon is nonconformal displacements of atoms of different sizes and masses, which causes a change in the shape of the Brillouin zones. The latter leads to a transformation of the Fermi surface and a change in the ratio of its hole and electron sheets.

CORRELATION OF THE CONVERSION PROCESSES WITH THE ANOMALY OF SPONTANEOUS MAGNETIZATION

In [6], a graph of the temperature dependence of spontaneous magnetization, $I_s(T)$, of annealed AlCoCuCrNiFe HEA is presented. Its temperature maximum, T_{max}~207 K, Fig. 2, coincides with the temperature of the input conversion (i.e. h→e) of our annealed $Al_{0.5}$CoCuCrNiFe samples, Fig. 1,a. Moreover, a decrease in T from T_{max} to ~170 K leaves I_s virtually unchanged, which correlates with the lack of conversion in our samples. Another interesting fact also attracts attention, namely, the dependence $I_s(T)$ below T~170 K (the region where part of the electron-like carriers are converted into holes) is characterized by a positive derivative, that contradicts all theoretical concepts [21]: ferromagnetic ordering of spin quanta should increase with decreasing T, but not vice versa. We will give a possible explanation for this contradiction, using results obtained above.

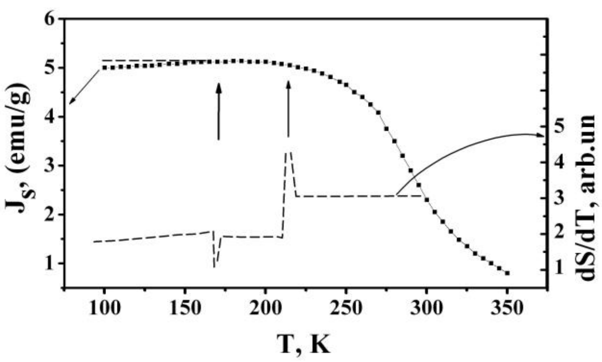

Fig. 2. Dependence Js(T) for AlCoCuCrNiFe [6] and schematic dependence of the derivative dS(T)/dT of our initial samples, Fig. 1,a

A key clue to the possible explanation in decrease of I_s below T~170 K is the e→h conversion occurring here. As result of the latter the number of N_e decreases. Or, in other words: an increase in the hole concentration causes a decrease in I_s. The picture looks as if the spontaneous magnetization I_s had identically directed electron and hole components, I_{se} and I_{sh}, respectively, and at the same time $i_{se}>i_{sh}$, where i_{se} and i_{sh} are the quanta of these components. In such case, the value $I_s=I_{se}+I_{sh}$ receives, as a result of the input (h→e) conversion, an addition proportional to the product of the number of converted holes and the difference $i_{se}–i_{sh}$. Since there is no conversion in the range of ~210…170 K, the value $I_s=I_{se}+I_{sh}$ should not change within the framework of the assumption made. And this is confirmed by the presence of a "plateau" on the $I_s(T)$ graph. After e→h, transformation at T~170 K, the I_s should decrease with temperature to the extent that the conversion additive is absent.

The question arises about validity of the assumption made.

Let us recall three main groups of the based models of ferromagnetism [21]. This is group of models of spin magnetism of conduction electrons (Ya.I. Frenkel, 1928), group of models of exchange interaction of rigidly localized ions of a crystal lattice (W. Heisenberg, 1928) and group of intermediate s-d models (S.P. Shubin, S.V. Vonsovskiy, 1936), combining both concepts. In all models, ferromagnetism is a consequence of the collinear ordering of effective quanta of spontaneous magnetization as a result of the exchange interaction between the carriers of these quanta. The undoubted correlation of our kinetic data with the magnetic anomaly [6] indicates that for description of the ferromagnetism of alloys AlCoCuCrNiFe family is most favorable one of the s-d models from third group. In other words: one needs taking into account both the spin and orbital magnetisms.

In a rough approximation, magnetization I_s of the system of charge carriers moving along of crystal lattice is proportional to their orbital momentum L, which in one's turn is proportional to mobility u: $I_s \propto |L| \propto u$. In presence of the carriers of both types and taking into account the relation $u_e>u_h$ established above, we obtain, in fact, confirmation of the above stated assumption. As for the relevance of the assumption that I_s has two components, one seems it naturally expands the list of physical characteristics (electrical conductivity, thermopower, thermal conductivity, Hall coefficient, etc.) adequately described within the framework of the phenomenological concept of two types of charge carriers.

CONCLUSIONS

1. It is shown that anomalies in the thermoelectric, electrical and thermal conductivity properties of $Al_{0.5}CoCuCrNiFe$ in the temperature range of ~250…77 K are associated with a change in the ratio between the number of electron-like and hole-like charge carriers due to hole↔electron conversion.

2. In the case of most thoroughly studied annealed structure, there is an interval of ~210…170 K at the entrance to which, from the high T side, a conversion of the hole→electron type occurs, leading to an increase in the area of the electronic part of the Fermi surface due to the area of the hole sections. When leaving this interval (T~170 K), there is a tendency to restore the original ratio of these areas due to conversion in the opposite direction (electron→hole).

3. In plastically deformed samples the temperature ranges of anomalies and directions of conversions at the upper and lower boundaries depend in a complex way on the degree of deformation ε. In particular, with increasing ε, the upper boundary of the anomaly shifts towards low temperatures.

4. In both annealed and deformed samples, an increase in the number of electron-like carriers is observed below ~145 K due to the hole→electron conversion. This factor is apparently responsible for the additional increase in strength of $Al_{0.5}CoCuCrNiFe$ with decreasing T.

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ВПЛИВ ПЛАСТИЧНОЇ ДЕФОРМАЦІЇ НА КОНВЕРСІЮ НОСІЇВ ЗАРЯДУ У ВИСОКОЕНТРОПІЙНОМУ СПЛАВІ $Al_{0,5}$CoCuCrNiFe

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У діапазоні ~300…77 K досліджено аномалії фундаментальних коефіцієнтів переносу високоентропійного сплаву $Al_{0.5}$CoCuCrNiFe, а також вплив пластичної деформації на ці аномалії. Як самі аномалії, так і деформаційні ефекти пов’язані, на нашу думку, з конверсією дірка↔електрон, що змінює співвідношення площ електронних і діркових листів поверхні Фермі. Кінетичні дані порівняно з температурною поведінкою спонтанної намагніченості (I_s) еквіатомного сплаву AlCoCuCrNiFe, що містить набір 3-d-феромагнетиків майже у тих же пропорціях. Інтерес до AlCoCuCrNiFe викликано наявністю аномалії $I_s(T)$ у тому ж діапазоні температур, де ми спостерігали кінетичні особливості $Al_{0,5}$CoCuCrNiFe. Запропоновано пояснення цієї магнітної аномалії явищем конверсії дірка↔електрон за припущенням, що і) I_s має електронну (I_{se}) та діркову (I_{sh}) компоненти і ii) кванти цих компонентів (i_{se} і i_{sh}) задовольняють вимозі $i_{se}>i_{sh}$.