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Low-temperature kinetic properties of the amorphous alloy $\text{Ca}_x\text{Al}_{1-x}$ ($0.2 < x < 0.8$)

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Abstract. Three electronic kinetic properties in dependence on temperature and alloy component concentration of amorphous alloy $\text{Ca}_x\text{Al}_{1-x}$ are calculated in the framework of the dynamic concentration excitation (DCE) model. It is shown that the electrical resistivity, thermopower and heat conductivity exhibit specific glassy anomalies susceptible to the change of alloy component concentration. This is corroborated by the experimental data.

1. Introduction

The lack of systematic experimental investigations of electron transport properties of non-magnetic amorphous alloys does not allow one to construct the common picture of electron transport in these materials [1–3]. There have been attempts to carry out the systematic analysis of the low-temperature property peculiarities in amorphous alloys, e.g. for $(\text{Ni}_{67}\text{X}_{33})_{1-x}\text{Al}_x$ ($\text{X} = \text{Ti}, \text{Zr}$ and La) [4] and Ca-Mg-Al metallic glasses [5], $\text{Zn}_x\text{Mg}_{1-x}$, $\text{Cu}_x\text{Sn}_{1-x}$ and $\text{Cu}_x\text{Al}_{1-x}$ [6–8], $\text{Ca}_x\text{Zn}_{1-x}$ [9] and $\text{Ca}_x\text{Al}_{1-x}$ [10]. The electrical resistivity ρ , thermopower S , Hall coefficient and magnetic susceptibility of these systems demonstrates the ‘glassy’ low-temperature behaviour of which the physical origin is still a problem for discussion. For example, the authors of [6] and [7] connect the observed anomalies in $S(T)$ (the non-linearity at low temperatures and the so-called ‘knee’ at $T = T_0$, where $T_0 \sim 20$ – 30 K) and in $\rho(T)$ (the negative temperature coefficient of resistivity at $T > T_0$) with phonon–roton states, but the low-temperature minimum in $\rho(T)$ is explained by the effect of electron–electron interaction (EEI) [11]. At the same time some results of [4–9] may be interpreted in the framework of a two-level system model or diffraction theory [1, 2].

The authors of [4], [5], [9] and [10] analysed the temperature and concentration dependences of resistivity from the point of view of the electronic structure of the alloys investigated. Their analysis of electron transport and electron structure in $\text{Ca}_x\text{Al}_{1-x}$ metallic glasses [10] attracted our attention. In fact, neither calcium nor aluminium involves d electrons in the form of the free atoms and therefore $\text{Ca}_x\text{Al}_{1-x}$ metallic glasses have been considered to be one of the simplest metallic glasses containing no transition metals. However, the electronic structure of these systems is shown to be not so simple [12]. Analysing photoemission spectra of $\text{Ca}_{0.7}\text{Al}_{0.3}$ and $\text{Ca}_{0.5}\text{Al}_{0.5}$ the authors of [12] found that the valence band is split into parts, taking this to be the breakdown of the free-electron-like band structure. They suggested that Ca d states emerge near the Fermi edge and that the d resonant effect causes a large resistivity.

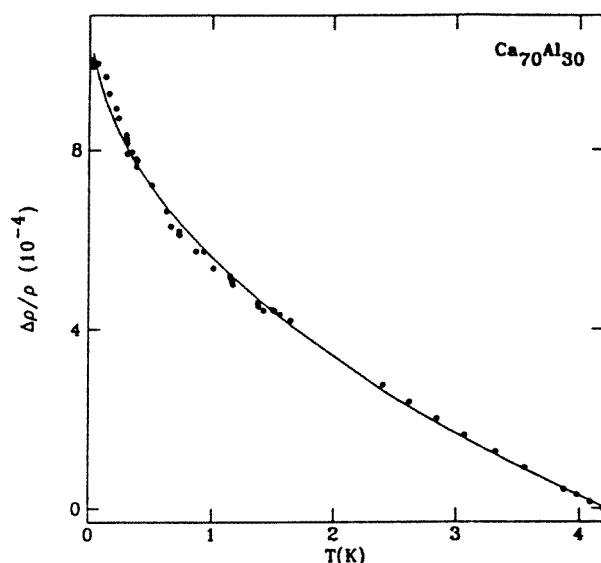


Figure 1. The low-temperature resistivity of $\text{Ca}_{0.7}\text{Al}_{0.3}$ [15]. The points are the experimental data and the solid line is a fit using the EEI model.

The data of the electronic specific heats, magnetic susceptibilities and Hall coefficients in $\text{Ca}_x\text{Al}_{1-x}$ [10] attributed a relatively large paramagnetism to the presence of the d-like states at the Fermi level. It was shown also that the electrical resistivity cannot be described in terms of the Ziman–Faber theory [1–3] and $\rho(T)$ is typical of the saturation type and can be expressed by the relation $\rho = \rho_0 - BT^{1.2}$ ($B > 0$) below about 120 K. The authors of this investigation [10] noted that the origin of the $T^{1.2}$ dependence of resistivity may be connected with the weak localization effect [13].

At the same time, in [14] and [15] it was shown that the nature of the low-temperature anomalous dependence of ρ is connected with the EEI effect and the weak-localization contribution is small and can be neglected. In figure 1 the low-temperature resistivity of $\text{Ca}_{0.7}\text{Al}_{0.3}$ is represented. The points are the experimental data and the solid line is a fit to the data [15], in good agreement with the experiment. This agreement is rather amazing, because it is obvious that the EEI theory developed for impurity metals [11] cannot be applied in its original form to amorphous alloys in which the concentrations of the components are of the same order of magnitude. In these systems the electrons are scattered not by a random impurity considered in [11] but rather by cluster-type structural non-uniformities. This is why to describe the electronic transport in metallic amorphous alloys the EEI theory [11] should be extended to the case of electronic scattering by structural formations of the short-range-order type. Such an extension was proposed in [16] where it was shown that the peculiarities of the low-temperature properties of non-magnetic amorphous metals are connected with the interference of inelastic EEI and multiple elastic electron scattering on the dynamic concentration excitations (DCEs) [16]. The DCEs are the excitations of the electron–ion system which are responsible for the cooperative non-diffusive rearrangement of local atomic configurations in amorphous metallic alloys.

In the DCE model the amorphous structure is represented as a quenched liquid, in which there can be a variety of short-range-ordered regions, many of which (with respect to the

type of short-range order) are not encountered in crystalline state of this system. In this case, the interaction potential of the amorphous system typically contains several minima of which the deepest (the principal minimum) corresponds to the crystalline state of the system and all other minima correspond to one type or another of short-range order. The short-range-ordered regions forming the secondary minima of the interaction potential, are the clusters, in which the interatomic bond determined by the spatial distribution of the electrons is different from the crystalline bond. This bond is of a dynamical (resonant) character and, correspondingly, it is formed by 'resonance' d (f) electrons. The short-range-ordered formations are maintained stationary by the fluctuation states they form [17]. The relaxation of the fluctuation electronic states to the energetically more favourable states results in the formation of a new chemical bond between atoms in a cluster (i.e. its rearrangement) and the creation of the DCEs [17].

It should be noted that the results of [16] for the electrical resistivity and electronic density of states agree with the corresponding results of the EEI theory [11] in the limit when the concentration of one of the alloy components $c \rightarrow 0$.

In [18] and [19] it was shown that the electron scattering on the DCEs, resulting in the renormalization of effective EEI vertex, is also a reason for the formation of the low-temperature anomaly of thermopower and electron heat conductivity.

Thus [16]–[19] showed that the peculiarities of the low-temperature behaviour of $\rho(T)$, $S(T)$ and $k(T)$ of non-magnetic amorphous alloys may have a common physical origin connected with the pure structure effects. This conclusion is substantiated by the results of [20]–[22] where the numerical calculations of $S(T)$ in amorphous $\text{Ca}_x\text{Al}_{1-x}$ ($0.55 \leq x \leq 0.80$) and $\text{Au}_x\text{Ni}_{1-x}$ ($0.15 \leq x \leq 0.80$) [20, 21], and also the calculations of $\rho(T)$, $S(T)$ and $k(T)$ in $\text{Au}_x\text{Ni}_{1-x}$ ($0.60 \leq x \leq 0.70$) [22] have been carried out. The comparison of the results for $S(T)$ in $\text{Ca}_x\text{Al}_{1-x}$ and for $\rho(T)$ in $\text{Au}_x\text{Ni}_{1-x}$ with the existing experimental data [23, 24] shows good qualitative and quantitative agreement.

Unfortunately in [22] we could not compare the results of the calculation of $S(T)$ and $k(T)$ with the corresponding experimental data for $\text{Au}_x\text{Ni}_{1-x}$ because of a lack of the latter. A similar situation with experimental data of electron transport properties is an inherent feature practically for all amorphous alloys. For example, for $\text{Ca}_x\text{Al}_{1-x}$ the thermopower and resistivity are well studied in the range $0.20 \leq x \leq 0.75$, but $k(T)$ has not been investigated.

The aim of the present paper is to study consistently the temperature and concentration dependences of electrical resistivity, thermopower and heat conductivity of amorphous alloys $\text{Ca}_x\text{Al}_{1-x}$ in a wide range of component concentration $0.20 < x < 0.80$.

The authors hope that the results obtained here and in [22] will attract the attention of physicists dealing with experiments in this range of physics of the amorphous state, and initiate the conduction of the corresponding investigations. The results of these experiments could answer the question of how adequately the DCE model would describe the influence of the self-structure state of amorphous alloys on the electron transport at low temperature.

2. A calculation of the electrical resistivity, thermopower and heat conductivity in amorphous alloy $\text{Ca}_x\text{Al}_{1-x}$

The DCE model may be applied only to amorphous metallic alloys with high electrical resistivity at low temperatures because the DCEs appear due to the d (f) resonance electrons forming new chemical bonds which lead to the formation of the new short-range-ordered regions, which are at low temperatures strong scattering centres for conduction electrons [18]. The amorphous alloy $\text{Ca}_x\text{Al}_{1-x}$ is a high-resistivity alloy, which is why we can use

our model for the description of its low-temperature electron transport.

The contribution made to the temperature dependence of electrical resistivity by the electron scattering processes mentioned above has the form

$$\delta\sigma(T)/\sigma \approx (2.5\sqrt{2}/6\pi^2\nu_0 D^{3/2})T^{1/2}/[1 + (T/T_0)^2]^{-1/4}. \quad (2.1)$$

Here ν_0 is the original electron density of states at the Fermi level, $D = v_F^2\tau/3$ is the electron diffusion coefficient and T_0 is the characteristic temperature of the amorphous alloy dividing the ranges of more and less intensive electron scattering on the DCEs [16]. It is defined as follows:

$$T_0 \approx (2\pi\hbar)^3 \mu_0^{1/2} \rho_0^2 \omega_0 / x^2 (1-x)^2 (\Delta E)^2 (2m)^{3/2} \kappa^3 \quad (2.2)$$

where μ_0 is the electron chemical potential, ρ_0 is the atomic density of the alloy, ΔE is the energy of alloy formation, $\kappa^2 = 4\pi e^2\nu_0$ and ω_0 is the boundary frequency of the DCEs. Estimation has shown that for amorphous alloys of $\text{Ca}_x\text{Al}_{1-x}$ type $\omega_0 \sim 100$ K [19, 20].

In the present work the electrical resistivity of six amorphous alloys $\text{Ca}_x\text{Al}_{1-x}$ for $x = 0.24, 0.35, 0.43, 0.50, 0.67$ and 0.78 is investigated. Calculating T_0 for these concentrations of Ca we took into account that the atomic density and formation energy depend on the alloy component concentration. The result of calculation of the contribution from the EEI and electron scattering on the DCEs to the electrical resistivity is represented in figure 2. As seen from figure 2, in the low-temperature range $T < 10T_0$ the resistivity decreases dramatically when temperature increases and then at $T_{sat} \sim 10T_0$ it goes to a 'plateau'. This result is in good qualitative agreement with the data of [10], [14] and [15].

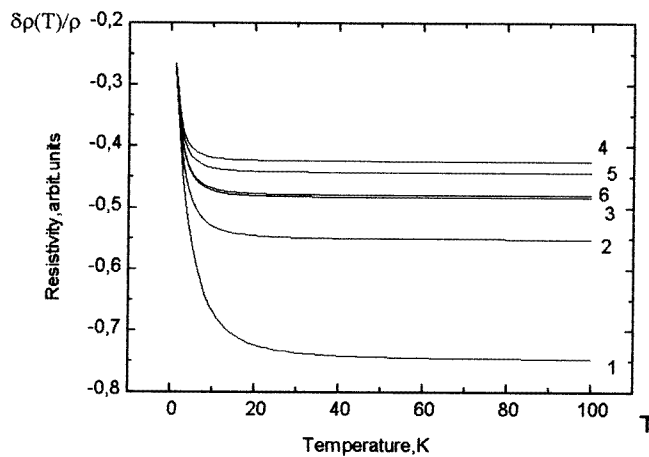


Figure 2. The correction to electrical resistivity of amorphous $\text{Ca}_x\text{Al}_{1-x}$ calculated taking into account the inelastic EEI and multiple elastic electron scattering by the DCEs for $x = 0.24$ (curve 1) 0.35 (curve 2), 0.43 (curve 3), 0.50 (curve 4), 0.67 (curve 5) and 0.78 (curve 6).

On can see from figure 2 that the depths of the $\rho(T)$ decrease and the disposition of the point of saturation depends on the component concentration of the alloy. In fact, an increase of Ca concentration from 24 to 67 at.% leads to a decrease of the depths and T_{min} . The subsequent increase of Ca concentration to 78 at.% results in an increase of the depths in $\rho(T)$ and a corresponding increase of T_{sat} .

Such a strong dependence of T_{sat} on component concentration of amorphous alloy $\text{Ca}_x\text{Al}_{1-x}$ is connected with the concentration dependence of the characteristic temperature

T_0 represented in figure 3. As seen from figure 3 T_0 has a minimum in the range of 55–65 at.% Ca and the dependence $T_0(x)$ is not symmetric about its minimum because of the dependence of the characteristic temperature on the formation energy and atomic density of the alloy, which also depend on alloy component concentration.

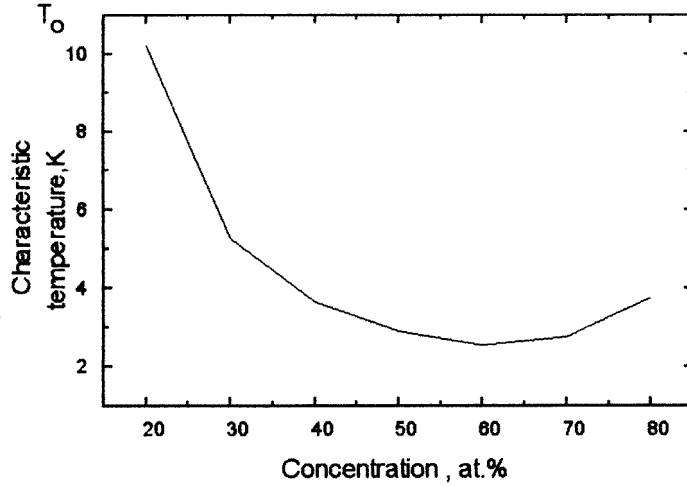


Figure 3. The characteristic temperature of amorphous alloy $\text{Ca}_x\text{Al}_{1-x}$ in dependence on concentration of Ca.

As will be shown below the dependence $T_0(x)$ represented in figure 3 leads to the shift of the ‘knee’ in $S(T)$ and the plateau in $k(T)$: their disposition is connected in the first case with $10T_0$ and in the second case with the range of temperature $T_0 < T < 10T_0$.

The thermopower of amorphous alloy $\text{Ca}_x\text{Al}_{1-x}$ connected with the interference of inelastic EEI and multiple elastic electron scattering by the DCEs is calculated in the following way [18–20]:

$$S(T) = (k_B^2 \pi^2 T / 3 |e|) \left[\tau_{eff}(\varepsilon) \frac{\partial}{\partial \varepsilon} \tau_{eff}^{-1}(\varepsilon) \right]_{\varepsilon=0} \quad (2.3)$$

where k_B is the Boltzmann constant, τ_{eff} is the effective relaxation time of the electrons and ε is the energy counting from the Fermi level.

The effective relaxation time of electrons taking into account the above-mentioned electron scattering processes is defined as follows [18]:

$$\tau_{eff}^{-1}(\varepsilon, T) = (m/2(2\pi)^3 v_0 \tau D^{3/2}) [1 + (T/T_0)^2]^{-1/4} J(\varepsilon, T) \quad (2.4)$$

where

$$J(\varepsilon, T) = \int_0^\infty \frac{dx}{\sqrt{x}} \left\{ \frac{N[(x + 1/2\tau - \mu_0)/T]}{\sqrt{x + 1/2\tau - \mu_0 - \varepsilon}} - \frac{N[(x + 1/2\tau + \mu_0)/T]}{\sqrt{x + 1/2\tau + \mu_0 - \varepsilon}} \right\} \quad (2.5)$$

and, finally, $N(x) = [\exp x - 1]^{-1}$.

The results of thermopower calculation for $\text{Ca}_x\text{Al}_{1-x}$ with $x = 0.24, 0.35, 0.43, 0.67$ and 0.78 are represented in figure 4. As seen from figure 4, the disposition of the low-temperature ‘knee’ in $S(T)$, as for the point of saturation in $\rho(T)$, depends strongly on the component concentration of the amorphous alloy. The ‘knee’ in $S(T)$ is located at $T_{knee} = 10T_0$, and T_0 first decreases (see figure 2) and then increases when Ca concentration increases. Hence the temperature T_{knee} will behave in the same way.

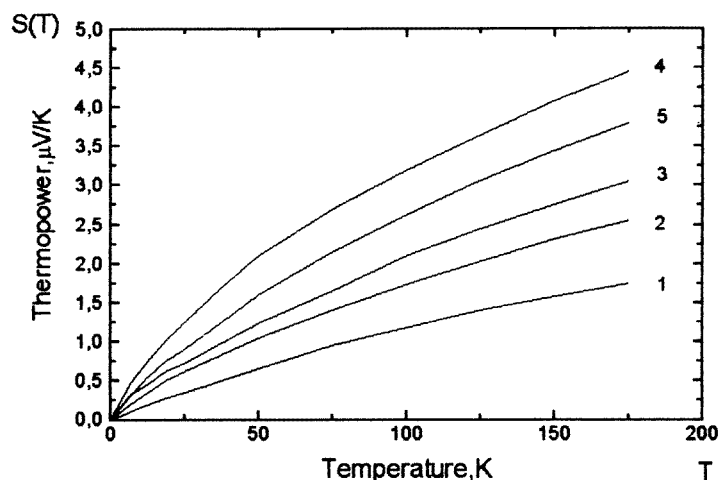


Figure 4. The calculated thermopower of amorphous alloy $\text{Ca}_x\text{Al}_{1-x}$ for $x = 0.24$ (curve 1), 0.35 (curve 2), 0.43 (curve 3), 0.67 (curve 4) and 0.78 (curve 5).

These results of the calculation of $S(T)$ in the amorphous alloy $\text{Ca}_x\text{Al}_{1-x}$ have been discussed in detail in our previous works [18,20,21] which is why we shall not dwell on this question here and note only that they agree well with the experimental data [23] and describe exactly the observed changes in low-temperature behaviour of thermopower connected with the variation of alloy component concentration.

It is well known that in metals the electron contribution to heat conductivity can be significantly greater than the phonon contribution [25], especially at low temperature. This is why it is interesting to calculate the electron heat conductivity in the amorphous alloy $\text{Ca}_x\text{Al}_{1-x}$ taking into account the interference of inelastic EEI and multiple elastic electron scattering by the DCEs. In [19] it was shown that the correction to the heat conductivity due to the mentioned electron scattering processes has the form

$$\delta k(T) = \left(5\sqrt{2}/36\pi^2 D^{1/2}\right) T^{3/2} \{1 + (T/T_0)^2\}^{-1/4} \Gamma(5/2) \zeta(5/2) \quad \text{at } T \ll T_0 \quad (2.6a)$$

$$\delta k(T) \approx [(1.83 \times 2^{5/2} \pi \kappa^2 D^{1/2})/24] T^{1/2} \Gamma(1/2) \quad \text{at } T_0 < T < 10T_0 \quad (2.6b)$$

$$\delta k(T) \approx (1.83 \pi \kappa^2 D^{1/2}/12) \Gamma(1/2) T_0^{3/2} T^{-1} \quad \text{at } T \gg 10T_0. \quad (2.6c)$$

The results of calculation of $\delta k(T)$ in amorphous $\text{Ca}_x\text{Al}_{1-x}$ for $x = 0.24, 0.35, 0.43$ and 0.67 at.% Ca are represented in figure 5. As seen from figure 5, at very low temperatures $T \ll T_0$ the heat conductivity behaves in the classical ‘glassy’, way [1–3], dramatically increasing (almost as T^2) when T increases, and then, in the intermediate-temperature range $T_0 < T < 10T_0$, going into a plateau. The disposition of the plateau depends strongly on the alloy composition. In fact, in the investigated concentration range the decrease of Ca concentration leads to a shift of the plateau to a higher temperature range, so for the alloy with the minimum characteristic temperature T_0 ($\text{Ca}_{0.67}\text{Al}_{0.33}$) the portion with the plateau is located on the temperature axis nearer to the beginning of the co-ordinates than for alloys with larger T_0 . It should be pointed out that the portion with the plateau will be also shifted to a higher temperature range if the Ca concentration increases from 0.67 at.% to 0.80 at.% because of the dependence of the characteristic temperature of the amorphous

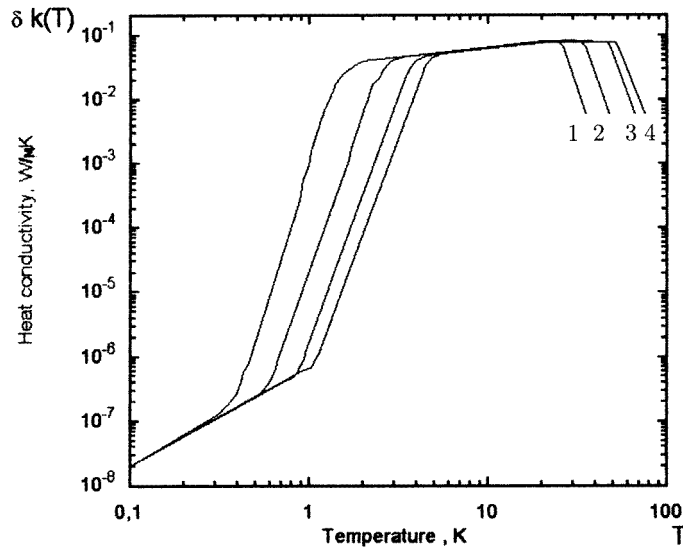


Figure 5. The contribution to the heat conductivity of $\text{Ca}_x\text{Al}_{1-x}$ from the interference of EEI and electron scattering on the DCEs for $x = 0.67$ (curve 1), 0.43 (curve 2), 0.35 (curve 3) and 0.24 (curve 4).

alloy $\text{Ca}_x\text{Al}_{1-x}$ on its concentration (see figure 3).

In the temperature range $T \gg T_0$ its contribution to electron heat conductivity calculated taking into account the interference of inelastic EEI and multiple elastic electron scattering by the DCEs decreases when T increases as T^{-1} , which is an inherent feature of metals with strong interelectron scattering [25]. However, at these temperatures the phonon mechanism of heat transport may come into play and predominate over the electron mechanism. That is why on the experimental curve $k(T)$ a portion with increasing heat conductivity may appear at $T \gg T_0$ [1–3].

3. Discussion

We have represented here the results of the simultaneous calculation of three electronic kinetic properties of the amorphous alloy $\text{Ca}_x\text{Al}_{1-x}$ in dependence on temperature and alloy component concentration. The electrical resistivity, thermopower and heat conductivity are shown to be characterized at low temperature by ‘glassy’ anomalies susceptible to change of alloy component concentration. The calculated thermopower and electrical resistivity are in good agreement with experimental data for $\text{Ca}_x\text{Al}_{1-x}$ [23, 10, 14, 15]. Unfortunately, experimental data for the heat conductivity in this alloy are not known to the present authors, which is why the question of how adequate its description is in the framework of the developed model of the DCEs is still open.

Based on the fact that the above-described anomalies of $\rho(T)$, $S(T)$ and $k(T)$ are characteristics of many non-magnetic metallic amorphous alloys [1–10], and on our results confirmed experimentally for $\text{Ca}_x\text{Al}_{1-x}$ and $\text{Au}_x\text{Ni}_{1-x}$ [23, 24], one may nevertheless conclude that the low-temperature anomalies of electron transport properties are due to pure structure effects and, evidently, connected with interference of inelastic EEI and multiple elastic electron scattering on the DCEs. This effect proves to be very significant in the range

of temperature $T < 10T_0$, where T_0 is the characteristic temperature of the amorphous alloy which determines the range of the intensive electron scattering on the DCEs [16–19].

The authors of [6] and [7], who investigated the anomalies of $\rho(T)$ and $S(T)$ in the intermediate range of temperatures in the framework of the phonon-roton states, also point out the existence of some characteristic temperature T_0 depending on the component concentration of amorphous alloys. This temperature divides the ranges of different behaviour of $\rho(T)$ and $S(T)$. In figure 6 the concentration dependence of T_0 for the amorphous alloy $\text{Cu}_x\text{Al}_{1-x}$ obtained from the thermopower ($T_0^{S(T)}$) and electrical resistivity ($T_0^{R(T)}$) data are represented. If one leaves aside the free approximation of $T_0^{S(T)}$ at $x = 0.40$ at.% Al then the dependence of the characteristic temperature T_0 on the alloy component concentration obtained in [6] and [7] is in good agreement with the concentration dependence of our T_0 (figure 3) multiplied by ten (we recall that the anomalies in $S(T)$ and $\rho(T)$ appear in the range $T < 10T_0$). This is especially clearly seen in the comparison of the concentration dependence of the characteristic temperature T_0 (figure 3) with $T_0^{R(T)}$ (figure 6).

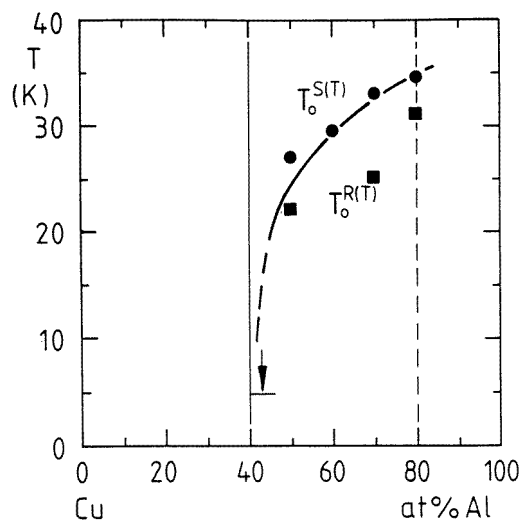


Figure 6. The characteristic temperature of the amorphous alloy $\text{Cu}_x\text{Al}_{1-x}$ obtained from the experimental data on thermopower ($T_0^{S(T)}$) and electrical resistivity ($T_0^{R(T)}$) in dependence on concentration of Al [6, 7].

We would like to comment here also on the results of the experimental investigations of the temperature dependence of the thermopower in non-magnetic metallic alloys published in [26] where the authors compared the dependence of $T_0^{S(T)}$ on the effective electron relaxation time calculated in [18] at low temperatures ($T < 200$ K) with the dependence of $T_0^{S(T)}$ on the resistivity at room temperature. Such a comparison seems to be incorrect because at room temperature the DCE contribution to the electrical resistivity is shown not to be significant [18] and it is possible to compare these dependences only at low temperatures. This is why correlation between $T_0^{S(T)}$ and $\rho_{300\text{K}}$ was not found in [26]; this cannot be taken as proof of the absence of the DCE effect on the low-temperature behaviour of thermopower of amorphous alloys.

We have shown here that there is a very strong dependence of T_0 on the alloy component concentration which may be investigated in experiments. We have shown also that the

electron scattering on the DCEs leads to the formation of the low-temperature anomalies of the electron transport properties of the amorphous alloy Ca_xAl_{1-x} and that the description of the low-temperature dependences of the electrical resistivity and thermopower in the framework of the DCE model is in good agreement with the experimental data.

Thus one may affirm that the concept of the dynamic concentration excitations gives confirmed results in the description of the low-temperature kinetic properties of non-magnetic amorphous metallic alloys. However the final estimation of these results may be made only after their comparison with the corresponding experimental data, the lack of which was pointed out at the beginning of the present work.

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