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SUPERCONDUCTIVITY AND ROOM TEMPERATURE COEFFICIENT OF RESISTANCE IN Zr-3d GLASSY ALLOYS

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Dedicated to Professor Mladen Paić on the occasion of his 90$^{\mathbf{th}}$ birthday

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The measurements of the superconducting transition temperatures T_c and the resistivity variation, including the room temperature coefficient of resistivity $\alpha = (\mathrm{d}\ln\rho/\mathrm{d}T)_{273}$, have been made for three binary Zr-3d (3d=Co, Ni or Cu) amorphous alloy systems and a number of ternary $\mathrm{Zr_2(Ni_{100-\mathit{x}}M_\mathit{x})_1}$ amorphous alloys (M=Ti, V, Cr, Mn, Fe, Co or Cu). In binary alloys, T_c decreases with decreasing Zr-content in agreement with the results of the photoemission studies and heat capacity measurements. However, the proportionality between the electron-phonon coupling constant λ (deduced from T_c) and $\mathrm{d}\rho/\mathrm{d}T$ predicted by the Ziman model is not generally valid. The variation of T_c and α in ternary alloys are strongly affected by the magnetic correlations for M = Cr and Mn. The observed $\alpha < 0$ for all alloys can be qualitatively explained in terms of incipient electron localization.

1. *Introduction*

This article is devoted to the electronic transport properties of amorphous transition metal–transition metal alloys at temperatures below the room temperature.

It was due to the effort of Professor Paić that the physics of the disordered (and metastable) metals and the low temperature physics have been introduced in Zagreb. In this paper, we report on the analysis of systematic measurements of the superconducting transition temperatures T_c and the resistivity variation of selected Zr-3d amorphous alloy systems. In particular, we studied the binary Zr-3d alloy systems $Zr_{100-x}Cu_x$ ($26 \leq x \leq 71$), $Zr_{100-x}Ni_x$ ($22 \leq x \leq 67$) and $Zr_{100-x}Co_x$ ($19 \leq x \leq 35$) and the ternary amorphous alloys $Zr_2(Ni_{100-x}M_x)_1$ (M=Ti, V, Cr, Mn, Fe, Co and Cu) with $x < 50$. In the earlier reports, Refs. 1 – 4, the temperature dependence of electrical resistivity and the concentration dependence of magnetic susceptibility have been discussed. We analyse in some detail the temperatures of the superconducting transition T_c and other parameters (such as $\alpha = \mathrm{d} \ln \rho / \mathrm{d}T$ that are closely related to the electronic band structure and the electron–phonon coupling in these alloy systems.

2. Experimental procedures

The amorphous binary and ternary Zr-3d alloys have been prepared by the melt spinning technique from the master alloys with the predetermined concentration, as described in Ref. 1. The samples in a form of ribbons were about 2 mm wide and 20–50 μm thick. The amorphous structure of the ribbons was verified by the X-ray diffraction.

The temperature dependence of electrical resistivity of the binary $Zr_{100-x}Cu_x$, $Zr_{100-x}Ni_x$ and $Zr_{100-x}Co_x$ alloys has been measured in the temperature range 1.5 K – 300 K, using the special cryostat immersed in liquid helium. The superconducting transitions were monitored resistively using the same setup. The actual resistivity measurements were performed with the potentiometric setup with the resistivity resolution of about 5 ppm.

Resistivities of the ternary $Zr_2(Zr_{100-x}M_x)_1$ alloys were measured in the temperature range 8 K – 300 K by using the cryogenic refrigerator (RMC-cryosystem LTS-22). The actual measurements were performed with standard four-probe either low frequency AC or DC techniques. The resolution in the resistivity measurements was about 10 ppm. The superconducting transition temperatures of these alloys were determined by the induction technique with samples immersed in liquid helium bath (pumped to the desired temperature).

3. Results and discussion

In the previous reports [3, 4] we discussed the variation in the temperature dependence of the electrical resistivity of amorphous Zr-3d alloys associated with the change in the concentration of 3d element. It was shown that over a broad temperature range variations of the conductivity in all these alloys can be qualitatively explained in terms of the incipient electron localization. In particular, the conductivity was found to increase approximately linearly with T at lower temperatures ($T \leq \Theta_D/3$) and as a $\sqrt{T}$ at somewhat higher temperatures ($T \geq \Theta_D/3$). The coefficients A of a $\sim T$ and B of $\sim \sqrt{T}$ variations of conductivity were associated with the strength of the electron–phonon interaction. In the binary amorphous Zr-Ni, Zr-Cu and Zr-Co alloys these coefficients decrease monotonically with decreasing Zr content, indicating that, as far as the electron–phonon interaction is concerned, the alloying of Zr with the late 3d elements can be understood in terms of the dilution effects. The main results of these measurements are given in Table 1: α is the room temperature coefficient of the electrical resistivity ($\mathrm{d}\ln\rho/\mathrm{d}T$ at 273 K), ρ_{273} the resistivity at the ice point and A is the coefficient of the linear variation of conductivity observed below 100 K. Figure 1 shows the variations of the superconducting transition temperatures T_c and α with the concentration of 3d element for the investigated alloys. These quantities are associated with the electron–phonon coupling. As seen from Fig. 1 and Table 1, the majority of alloys is superconducting with T_c below 4 K. The superconducting transitions were quite sharp, with the width $\Delta T_c \leq 50$ mK. Within the explored concentration range, T_c decreases approximately linearly with 3d content in all three alloy systems. The behaviour of α of Zr-Ni and Zr-Co alloys is quite similar to that of T_c. In Zr-Cu alloys, there is a weak change of α for $x < 50$ (note that $\alpha < 0$ for all alloys). The average slopes $\Delta T_c/\Delta x$ (Fig. 1) are –0.114, –0.064 and –0.094 K/at% for Zr-Co, Zr-Ni and Zr-Cu alloy systems, respectively.

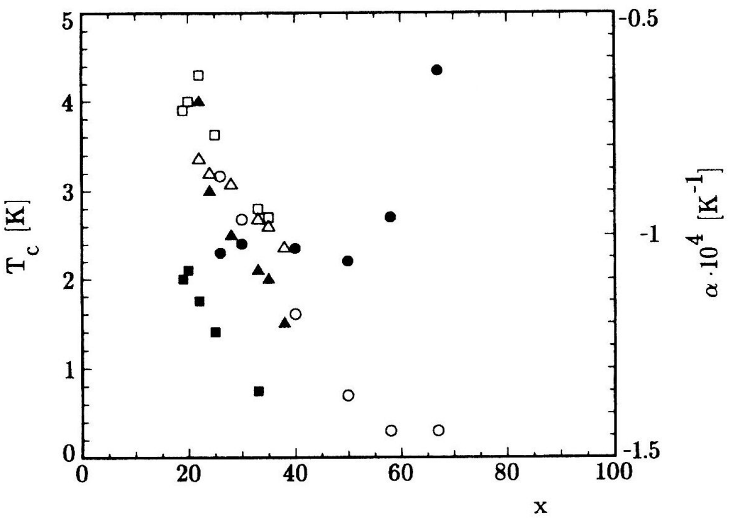

Fig. 1. Left scale: The temperature of the superconductivity transition T_c vs. concentration x of the 3d element for ZrCo *alloys □, ZrCu alloys ○ and ZrNi alloys △. Right scale: The room temperature coefficient of resistance α vs. concentration x of the 3d element for ZrCo alloys □, ZrCu alloys ○ and ZrNi alloys △.*

Since the scattering in transition metals and the electron–phonon coupling is strong, it was necessary to modify the BCS theory of the superconductivity in these systems. McMillan [5] has redefined the electron–phonon coupling constant λ for the superconductors with strong electron–phonon coupling and obtained the following relation for T_c:

$$T_c = \frac{\Theta_D}{1.45} \exp \frac{-1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}, \tag{1}$$

where Θ_D is the Debye temperature and μ^* is the effective matrix element of the interelectronic Coulomb interaction (estimated to be about 0.13). The electron–phonon constant λ can be expressed as:

$$\lambda = \frac{N(E_F) < I^2 >}{M < \omega^2 >_m}, \tag{2}$$

where $< I^2 >$ is the square of the electronic matrix element associated with the change of the crystal potential as an atom is moved and $< \omega^2 >_m$ is the average squared phonon frequency defined by McMillan [5]. The relation (2) can be written in a form $\lambda = N(E_F)\delta$ where $\delta =< I^2 > /M < \omega^2 >_m$. Varma and Dynes [6] have shown that for the transition metal alloys, δ is approximately constant.

TABLE 1.

Data relevant to $Zr_2(Ni_{100-x}M_x)_1$ *glassy alloys:* α *is the room temperature coefficient of resistance,* T_c *is the superconductivity transition temperature,* ρ_{273} *is the electrical resistivity at* 273 K *and* A *is the coefficient of linear conductivity variation below* 100 K.

	$\alpha \times 10^4$ (K^{-1})	T_c (K)	ρ_{273} ($\mu\Omega$cm)	$A \times 10^6$ ($\mu\Omega$cm)$^{-1}$
$Zr_{74}Cu_{26}$	–1.04	3.17		
$Zr_{70}Cu_{30}$	–1.02	2.68	160.6	1.31
$Zr_{60}Cu_{40}$	–1.03	1.60	164.9	1.36
$Zr_{50}Cu_{50}$	–1.06	0.7	167.6	1.13
$Zr_{42}Cu_{58}$	–0.96	0.3	167.0	0.9
$Zr_{33}Cu_{67}$	–0.63	0.3	162.8	0.68
$Zr_{76}Ni_{24}$	–0.90	3.20	165.6	0.9
$Zr_{72}Ni_{28}$	–1.00	3.06	166.9	1.20
$Zr_{67}Ni_{33}$	–1.08	2.68	172.4	1.31
$Zr_{65}Ni_{35}$	–1.10	2.60	174.2	1.31
$Zr_{62}Ni_{38}$	–1.20	2.36	173.5	1.31
$Zr_{37}Ni_{63}$	–0.50	1.2	176.2	0.92
$Zr_{81}Co_{19}$	–1.10	3.90	162.2	1.68
$Zr_{75}Co_{25}$	–1.22	3.63	162.3	0.82
$Zr_{67}Co_{33}$	–1.35	2.80	175.9	0.74
$Zr_{65}Co_{35}$	–2.00	2.70	182.5	1.35
$Zr_2(Ni_{50}Cu_{50})_1$	–1.17	2.41	170.2	1.28
$Zr_2(Ni_{90}Co_{10})_1$	–1.21	2.76	171.8	1.33
$Zr_2(Ni_{50}Fe_{50})_1$	–1.15	1.59	168.7	1.28
$Zr_2(Ni_{90}Mn_{10})_1$	–0.93	< 1.3	166.7	1.05
$Zr_2(Ni_{95}Cr_5)_1$	–1.14	1.56	173.5	
$Zr_2(Ni_{90}V_{10})_1$	–1.22	2.48	165.5	1.30
$Zr_2(Ni_{80}Ti_{20})_1$	–1.24	3.06	167.3	1.40

The investigations of the electronic band structure of the binary Zr-3d amorphous alloys by means of spectroscopic methods [7] have shown two distinct components in the valence band: one is close to the Fermi energy, E_F, and the other at higher binding energies. The shallow maximum close to E_F is associated to the Zr 4d-states, whereas a quite sharp maximum at higher binding energy belongs to d-states of the 3d element. The difference in the binding energies of 4d-states and 3d-states increases on going towards the end of 3d series (Cu). This seems to facilitate the interpretation of the properties of Zr-3d amorphous alloys with late 3d elements (Co, Ni, Cu).

Superconductivity in amorphous metals is often discussed in terms of the dependence of T_c on the number of electrons per atom (e/at), [8]. However, it was shown in Ref. 9 that due to reasons mentioned above this cannot be applied for the glassy Zr-3d alloys with the late 3d elements. In particular the variation of T_c with x in these alloys depends strongly on the positions of two d-subbands (4d and 3d, respectively) in the electronic density of states. An analysis of $Zr_{70}X_{30}$ amorphous alloys in Ref. 9, were X=3d or 4d element has shown that the relation (2) is approximately applicable to these alloys and that λ increases approximately linearly with $N(E_F)$. Using the McMillan relation (1) and measured T_c and Θ_D (from the low temperature heat capacity measurements) one can determine λ. In order to estimate λ, we used for the Coulomb pseudopotential $\mu^* = 0.13$, and the results from the heat capacity measurements on Zr-Cu alloys from Refs. 10 and 11 and Zr-Ni alloys from Ref. 12

The variations of the calculated λ and the measured coefficients A (from the low temperature linear conductivity variation) with the concentration x of the 3d elements for the investigated Zr-3d glassy alloys are shown in Fig. 2. Both λ and A are seen to decrease with increasing x (lowering of Zr content).

The Ziman's theory [13] predicts a linear relation between λ and $\mathrm{d}\rho/\mathrm{d}T$:

$$\lambda = k\frac{\mathrm{d}\rho}{\mathrm{d}T}, \tag{3}$$

where $\mathrm{d}\rho/\mathrm{d}T = \alpha\rho_{273}$. Although the experimental variation of λ with $\mathrm{d}\rho/\mathrm{d}T$ for Zr-Cu and Zr-Ni alloy can be roughly approximated with the linear one ($k = -28$ $(\mathrm{n}\Omega\mathrm{cm})^{-1}\mathrm{K}$), some data points show considerable deviation from Eq. (3). This is particularly true for Zr-Cu alloys. There may be several reasons for the observed deviations from the predicted variation described by Eq. (3): the alloys from the edges of the 3d concentration range may not be fully amorphous, the selected fixed value $\mu^* = 0.13$ may not be quite appropriate for all alloy systems as shown in Ref. 14, the spin–fluctuations may affect T_c (hence λ) of some alloys and, finally, for the alloys with very low T_c, the uncertainty in the actual value of T_c is quite large. However, since the Ziman's theory was not able to describe properly the temperature dependence of the resistivity (including $\mathrm{d}\rho/\mathrm{d}T$) in the investigated alloy systems, as shown in Ref. 2, the approximate proportionality between λ and $\mathrm{d}\rho/\mathrm{d}T$ cannot be taken as an evidence for the applicability of this model in the present case.

As pointed out earlier, for all investigated alloys, $\alpha < 0$ is observed. In particular (see Table 1), α varies from -0.6×10^{-4} K^{-1} to -1.24×10^{-4} K^{-1} in our alloys. The observed $\alpha < 0$ is consistent with the Mooij correlation [15], showing that in a number of alloys $\alpha < 0$ if $\rho \geq \rho_m = 150$ $\mu\Omega$cm.

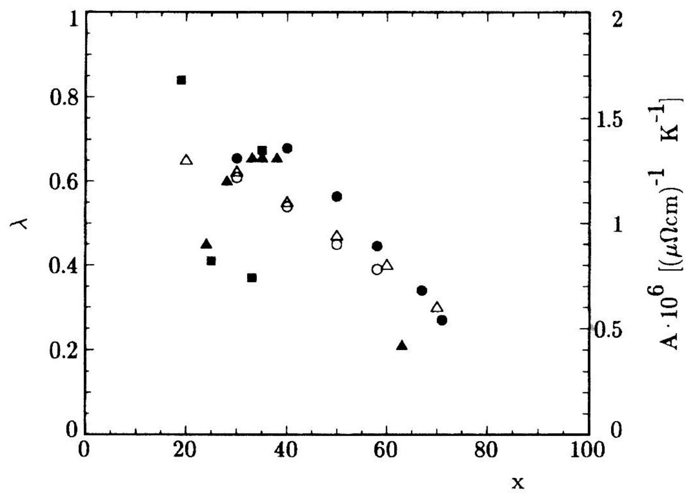

Fig. 2. Left scale: The coefficient of electron–phonon interaction λ vs. concentration x of M (M = Cu, Ni, Co) *for* ZrCu *alloys* ○, *and* ZrNi *alloys* △. *Right scale: Coefficient A of linear conductivity variation below* 100 K *vs. concentration x of* M *for* ZrCo *alloys* □, ZrCu *alloys* ○ *and* ZrNi *alloys* △.

Although the Ziman's theory predicts a definite relation between α and ρ, it does not predict the universal value of ρ_m above which $\alpha < 0$ should appear. Furthermore, the Ziman theory was not able to account for the magnitudes and signs of α throughout the explored range of x in amorphous Zr-3d alloys, as shown in Ref. 16. The appearance of $\alpha < 0$ at elevated resistivities and the observed temperature variations of conductivity can be associated with the effects of the incipient electron localization as shown by Kaveh and Mott [17]. The magnitude of the electrical resistivity can be calculated in terms of the classical ("Boltzmann's") theory (such as the extended Ziman model or the Mott model), whereas the explanation of the temperature variation of the electrical conductivity requires the use of the electron localization theory.

The Ziman's and Mott's models take into account the contribution of s-electrons to the conductivity only. However, in general the conductivity of transition metals consists of two terms:

$$\sigma = \sigma_s + \sigma_d, \tag{4}$$

where σ_s is the contribution due to s-band and σ_d is that due to d-band. Since the effective mass of d-electrons is quite large, their contribution to the conductivity in crystalline transition metals is negligible. However since in the amorphous transition metal alloys, the electronic mean free path is short and the number of s-states lowered due to s-d hybridization effects, σ_s and σ_d may become comparable. The measurements of the change in electrical resistivity of amorphous Zr-Cu and Zr-Ni alloys caused by the elastic deformation enabled the indirect estimation of σ_d. The σ_d/σ ratio, estimated by Marohnić et al. [18], seems to confirm the hypothesis on the significant contribution of d-electrons to the total conductivity of amorphous Zr-3d alloys.

Although the incipient localization provides a qualitative explanation of the negative temperature coefficient of resistivity in Zr-3d amorphous alloys, in order to obtain the quantitative results, Howson and Morgan [19] performed the calculations in terms of the two band model taking into account the effects of the s-d hybridization. In this approach, σ_s and σ_d include corrections performed due to the s-d hybridization. The calculated σ_s seems to be proportional to $N_d^{-1}(E)$, where $N_d(E)$ is the density of states calculated by Krajči [20] for amorphous and liquid Fe. One can estimate $\partial \ln N(E)/\partial T \approx 10^{-4}$. Therefore, one would expect a small negative contribution to resistivity associated with the temperature variation of σ_s. Compared with σ_s, the contribution of σ_d to the variation of the electrical resistivity seems negligible. Therefore, it seems that the temperature dependence of the electrical resistivity in these alloys is associated with the hybridized s-band. However, both σ_d and σ_s have to be taken into account in order to account for the measured magnitude of the conductivity.

Interesting conclusions about the electronic structure and correlations in these systems can be obtained by alloying the binary Zr-3d alloy system $Zr_2(Ni_{100-x}M_x)_1$, M being any of the 3d transition metals. The study of Ultraviolet Photoemission Spectroscopy (UPS) He I spectra and low temperature heat capacity, performed on ternary amorphous $(Zr_{67}Ni_{33})_{85}M_{15}$ alloys (M = Ti, V, Cr, Mn, Fe, Co, Ni or Cu) [21,22], showed a sizeable and systematic change of the electronic density of states caused by M. The UPS He I spectrum of amorphous Zr_2Ni alloy shows two maxima; one close to E_F is associated with Zr 4d-states and the other at the binding energy 1.8 eV below E_F belongs to Ni 3d-states. The addition of M causes a continuous increase of $N(E_F)$ in going from Cu towards Ti. In particular, the addition of Ti enhances $N(E_F)$ and reduces the intensity of the maximum associated with the 3d-states of Ni. When M = Cu, the Cu 3d-states form separate maximum at the binding energy of 3.5 eV. Accordingly, the addition of Cu reduces the $N(E_F)$ with respect to that of Zr_2Ni. In this respect the observations for the ternary alloys are consistent with the previous ones for binary Zr-3d alloys which also showed the continuous decrease of $N(E_F)$ in going towards the end of 3d series (Cu).

The variation of the coefficient of the electronic contribution to the heat capacity γ with M is in general agreement with the variation of $N(E_F)$ deduced from photoemission studies as shown in Ref. 21, except for the alloys with M = Cr and Mn which show the strongly enhanced γ. This behaviour can be attributed to the magnetic contribution (γ_m) to γ. The magnetic correlations for M = Cr and Mn also suppress strongly the superconductivity in these alloys. Figure 3 shows the gradient of the change of the superconducting transition temperature $\Delta T_c/\Delta x$ and the coefficient A vs. M (M=Ti, V, Cr, Mn, Fe, Co, Ni or Cu). Since in all alloys T_c

varied approximately linearly with x, this gradient represents the best effect of M.

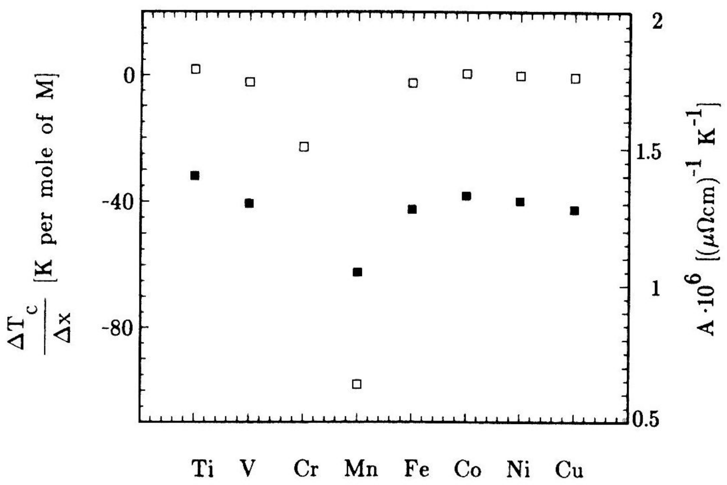

Fig. 3. The gradient of the change in the superconducting transition temperature $\Delta T_c/\Delta x$ *vs.* M *(member of 3d series) for the* $Zr_2(Ni_{100-x}M_x)_1$ *glassy alloys* □. *Right scale: coefficient A of variation of linear conductivity below* 100 K *vs.* M □.

The coefficients A in $Zr_2(Ni_{100-x}M_x)_1$ for a fixed (or similar) x showed a tendency to increase in going from Cu towards Ti with the superimposed rather deep minimum centered at M = Mn. This depression of A (associated with the electron–phonon coupling) is consistent with the strong magnetic contribution γ_m in the γ of alloys with Mn. The variation of $\Delta T_c/\Delta x$ with M is very similar to that of A. In particular $\Delta T_c/\Delta x$ increases little in going from Cu towards Ti. This is to be expected since both $\Delta T_c/\Delta x$ and A depend on the strength of the electron–phonon coupling and the density of states at the Fermi level. Our observations and the above discussion are, therefore, consistent with the results of UPS studies with He I ($h\nu = 21.2$ eV) and the heat capacity measurements performed on similar alloys [21, 22]. It is interesting to note that α (determined around 273 K) shows some increase in going from M = Cu towards M = Ti. Furthermore, α also shows pronounced minimum around M = Mn. This indicates that the changes in $N(E_F)$ and the electron–phonon coupling also determine the variation of α with M. However the simple relation (3) derived from the Ziman's theory is not sufficient in order to account for the observed behaviour.

4. Conclusion

The superconducting transition temperatures T_c and the coefficients A of the low temperature linear conductivity variation decrease linearly with decreasing Zr content in amorphous Zr-3d alloys. These variations are consistent with the results of the UPS and the low temperature heat capacity studies which provide an insight into the electronic structure of these alloys. In particular, the decrease of the Zr content lowers the electronic density of states at the Fermi level $N(E_F)$ and is associated with the decrease of the effective electron–phonon coupling. The decrease of T_c is related to the electron–phonon coupling constant λ given by the McMillan [5] expression (1). The effects of λ on the electrical resistivity (ρ) can be associated either with the incipient electron localization (via the inelastic scattering time) or within the framework of the "classical" Ziman's theory. Whereas similar variations of A and λ in the investigated Zr-3d alloys (3d = Co, Ni, Cu) provide some support for the incipient electron localization, the linear relation between λ and $(\mathrm{d}\rho/\mathrm{d}T)_{273}$ predicted by the Ziman model is observed in Zr-Ni alloys only.

The addition of the third transition element M (Ti, V, Cr, Mn, Fe, Co or Cu) to the amorphous Zr_2Ni alloy causes, for the fixed M content, a systematic decrease of T_c and A in going from M = Ti towards M = Cu. These findings are also consistent with the results of the UPS studies from Refs. 21 and 22, performed on similar alloys and showing the decrease in $N(E_F)$ on going from Ti towards Cu. The pronounced maximum in the rate of suppression of T_c by M ($\Delta T_c/\Delta x$) and A for M = Cr and Mn can be associated with the appearance of the magnetic correlations in the alloys containing these impurities, and is consistent with the observed enhancement of the coefficient of the electronic specific heat in similar alloys [21]. The coefficients A in ternary alloys can also be associated with the incipient electron localization. The proportionality between λ and $(\mathrm{d}\rho/\mathrm{d}T)_{273}$ is not fulfilled for ternary alloys.

For all investigated alloys the negative temperature coefficients of resistivity can be qualitatively explained in terms of the incipient electron localization in the high resistivity alloys. For the quantitative estimate of the magnitude of resistivity of these alloys, one may use the results of calculations performed in the two band model [19]. Combining these results with the recent calculation of the electronic density of states in amorphous and liquid Fe, as shown in Ref. 20, one may associate the changes in the temperature coefficient of resistivity α with the corresponding changes in the electronic density of states.

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SUPRAVODLJIVOST I TEMPERATURNI KOEFICIJENT ELEKTRIČNOG OTPORA NA SOBNOJ TEMPERATURI U Zr-3d STAKLASTIM SLITINAMA

Mjereni su temperatura supravodljivog prijelaza T_c i promjena električnog otpora s temperaturom kao i temperaturni koeficijent električnog otpora na sobnoj temperaturi $\alpha = (\mathrm{d}\ln\rho/\mathrm{d}T)_{273}$ za tri binarna Zr-3d (3d = Co, Ni ili Cu) staklasta sistema i za niz $Zr_2(Ni_{100-x}M_x)_1$ staklastih slitina (M = Ti, V, Cr, Mn, Fe, Co ili Cu). U binarnim se slitinama T_c smanjuje sa smanjenjem Zr i to je potpuno u skladu s rezultatima eksperimenata fotoemisije i mjerenja toplinskog kapaciteta dobivenim na sličnim slitinama, koji pokazuju da se elektronska gustoća stanja na Fermijevoj razini smanjuje sa smanjenjem količine Zr. Proporcionalnost između konstante elektron–fonon interakcije λ (izračunate iz T_c) i $\mathrm{d}\rho/\mathrm{d}T$, kako to predviđa Zimanov model, općenito ne vrijedi. Promjene T_c i α u ternarnim slitinama za slitine s M = Cr i Mn pod jakim su utjecajem magnetskih efekata. Uočene $\alpha < 0$ za sve slitine mogu se kvalitativno objasniti efektom početne lokalizacije elektrona.