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ELECTRONIC AND MAGNETIC PROPERTIES OF TERNARY $Zr_2(Ni_{1-x}M_x)_1$ GLASSY ALLOYS

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The electric resistivities (in the temperature range from 8 K to 300 K), the room temperature magnetic susceptibility and the superconducting transition temperature of the amorphous $Zr_2(Ni_{1-x}M_x)_1$ alloys (M = Ti, V, Cr, Mn, Fe, Co, Ni and Cu) have been investigated. The temperature dependence of the electrical resistivity can be described in terms of the incipient electron localisation effects inherent to the high resistivity alloys, same as of the other Zr-3d glassy alloys (3d = Fe, Co, Cu, or Ni). The new features introduced with M are the pronounced nonmonotonic variations of the magnetic and electron-transport properties with M. The origin of these variations are the systematic changes in the electronic band structure of the alloys on going from M = Cu towards M = Ti (as confirmed by the photoemission experiments on similar $(Zr_{67}Ni_{33})_{85}M_{15}$ amorphous alloys) and the tendency to the formation of localized magnetic moments for M around the middle of 3d-series (M = V, Cr, Mn, and Fe). A novel feature is a rather strong suppression of the effects of the incipient localization in the resistivity by the magnetic interactions.

1. Introduction

Previously we have reported the results of a detailed investigation of the electronic transport and magnetic properties of selected amorphous Zr-3d alloys [1,2]. In these studies we have focused our attention to the alloys containing late 3d elements, in particular Cu, Ni, Co and Fe. A smooth and quasi-linear variations of the investigated properties with 3d metal content suggested an interpretation in terms of the dilution effects. Such an interpretation was supported by the results of the photoemission experiments which showed [3] that the Zr-d band extends to the Fermi energy (E_{F}) whereas the 3d states of the above mentioned elements have much larger binding energies than the Zr-4d states. However, the binding energies of the 3d states are found to decrease on going from Cu to Fe. Therefore it seemed interesting to investigate the amorphous Zr-based alloys with 3d elements appearing before Fe in 3d series.

Since bulk amorphous alloys of Zr with these elements cannot be made we have extended our investigation to the ternary Zr-rich alloys. In particular we have investigated the amorphous $\mathrm{Zr_2(Ni_{1-x}M_x)_1}$ with M = Ti, V, Cr, Mn, Fe, Co, Ni and Cu, respectively. The preliminary measurements of the magnetic susceptibility and the superconducting transition temperature were in general agreement with the findings of the photoemission experiments but also indicated the formation of the local moments and/or the presence of the localized spin fluctuations for the addition of the metals close to the center of the 3d series [4]. In particular a maximum in the magnetic susceptibility and the minimum in T_c have been observed for M = Mn. More recent results of the low temperature heat capacity measurements support this conclusion. Here we present the results of the recent transport property measurements on these alloys and compare these results with the above findings for the thermodynamic properties.

2. Experimental procedure

The amorphous $\mathrm{Zr_2(Ni_{1-x}M_x)_1}$ alloys have been prepared by the melt spinning of the master alloys with predetermined concentration onto a rim of the rapidly rotating copper roller [4]. The resulting ribbons were about 2 mm wide and 30-50 μm thick. The amorphous state of the alloys was verified using the X-ray diffraction. For almost all $\mathrm{Zr_2(Ni_{1-x}M_x)_1}$ alloys the amorphous state has been obtained over sizeable range of x. The exception was M = Cr for which we were not able to obtain amorphous alloys with $x > 0.05$.

The magnetic susceptibility has been measured with the Faraday balance on samples with a mass of few milligrams. The temperatures of the superconducting transition were measured with the induction technique at 70 Hz with sample immersed in liquid helium. The superconducting transition temperatures T_c were obtained by controlled pumping of the cryostat. The electrical resistivities were measured on 3-5 cm long samples supplied with the thin Pt voltage and current leads. A standard four-point DC method with the sensitivity $1:10^4$ has been used.

TABLE 1.

	α	ρ_{273}	T_c	$\chi \cdot 10^3$	$\Delta\chi \cdot 10\,(JT^{-2}$	$A \cdot 10^6$	$B \cdot 10^5$
	K^{-1}	($\mu\Omega$cm)	K	($JT^{-2}mol^{-1}$)	(per mol of M)	$(\mu\Omega cm)^{-1}K^{-1}$	$(\mu\Omega cm)^{-1}K^{-1/2}$
Zr_2Ni	−1.08	172.4	2.68	1.178		1.31	2.36
$Zr_2(Ni_{0.5}Cu_{0.5})_1$	−1.17	170.2	2.41	1.096	−4	1.28	2.65
$Zr_2(Ni_{0.9}Co_{0.1})_1$	−1.21	171.8	2.76	1.329	28	1.33	2.56
$Zr_2(Ni_{0.5}Fe_{0.5})_1$	−1.15	168.7	1.59	1.626	93	1.28	2.50
$Zr_2(Ni_{0.9}Mn_{0.1})_1$	−0.93	166.7	<1.3	1.534	356	1.05	1.95
$Zr_2(Ni_{0.95}Cr_{0.05})_1$	−1.14	173.5	1.56	1.252	172		
$Zr_2(Ni_{0.9}V_{0.1})_1$	−1.22	165.5	2.48	1.281	92	1.30	2.44
$Zr_2(Ni_{0.8}Ti_{0.2})_1$	−1.24	167.3	3.06	1.313	68	1.40	3.00

Data relevant to $Zr_2(Ni_{1-x}M_x)_1$ glassy alloys; α is the room temperature coefficient of resistance, ρ_{273} is the electrical resistivity at 273 K, T_c is the superconductivity transition temperature, χ_p is the magnetic susceptibility at room temperature, $\Delta\chi$ is the change in magnetic susceptibility per mole of M, A is the coefficient of linear variation of conductivity variation bellow 100 K, B is the coefficient of a $T^{1/2}$ variation of conductivity above 100 K.

The temperatures in the range 8-300 K have been obtained with the cryocooler (Model RMC-Cryosystem LTS-22). Due to the small thickness of the samples the absolute values of resistivity were accurate to within few percents. Some data relevant to the investigated samples are given in Table 1.

3. *Results and discussion*

As mentioned in the Introduction we were particulary interested in the variation of the electrical resistivity of the investigated alloys with temperature. Since the resistivity ρ of all our alloys (Table 1) were high, and therefore the corresponding electronic mean free paths short, one would expect the presence of the quantum corrections to conductivity [5,6] as was observed in binary Zr-3d amorphous alloys [1,2]. Within the explored temperature interval ($T > 10\,\mathrm{K}$) the correction to conductivity due to incipient localisation [7] is expected to dominate the variation of conductivity with temperature. This contribution to conductivity is proportional to $(D\tau)^{-1/2}$, where D is the diffusion constant and τ is the characteristic phase coherence time. τ is expected to be determined by the inelastic scattering time τ_i which for the disordered nonmagnetic alloy should be related to the electron-phonon interaction. As discussed elsewhere [2], at temperatures above the Debye temperature Θ_D (in practice for $T > \Theta_\mathrm{D}/3$) $\tau_i^{-1} \sim T$ is expected. At lower, but not too low temperatures $\tau_i^{-1} \sim T^2$ is envisaged [8]. Accordingly, if one ignores the contributions other than that due to electron-phonon interaction, one expects to observe a linear variation of conductivity with T at lower and $T^{1/2}$ at higher temperatures, respectively. The results [1,2] for Zr-3d alloys (3d = Fe, Co, Ni and Cu) agreed qualitatively with those predictions. Moreover, the coefficients of the T and $T^{1/2}$ terms in conductivity showed the variations with 3d metal content which are consistent with the expected variations in the electron-phonon coupling. However, in order to obtain a semi-quantitative agreement between the experimentally observed conductivity variations and the model predictions [7] one should also take into account the spin-orbit interaction [9] (via τ_{s0}) which is quite strong in these transition metal alloys. Since at present we do not have a realiable information on the spin-orbit coupling in ternary $\mathrm{Zr_2(Ni_{1-x}M_x)_1}$ alloys, we shall proceed with the simplified analysis (ignoring τ_{s0}) outlined above and previously performed for amorphous Zr-3d alloys. As some justification for such an analysis we note that we do not expect larger variation of the τ_{s0} for rather small contents (x) of M in the investigated $\mathrm{Zr_2(Ni_{1-x}M_x)_1}$ alloys. Therefore, the observed variation should at least in a qualitative way reflect the variation in τ_i.

Figure 1 shows the temperature dependent contributions to conductivity for several amorphous $\mathrm{Zr_2(Ni_{1-x}M_x)_1}$ alloys. The change in conductivity is $\Delta\sigma = \sigma_T - \sigma_0$, where σ_T is the conductivity at temperature T and σ_0 is the extrapolation of the conductivity from the temperature interval in which it varies linearly with temperature to $T = 0$ K. Figure 1a shows the variation of $\Delta\sigma$ with T for $T < 100$ K in a linear temperature scale whereas Fig. 1b shows the variation of $\Delta\sigma$ for $T < 300$ K in a plot suitable to deduce the eventual $T^{1/2}$ variation. In spite of the expected

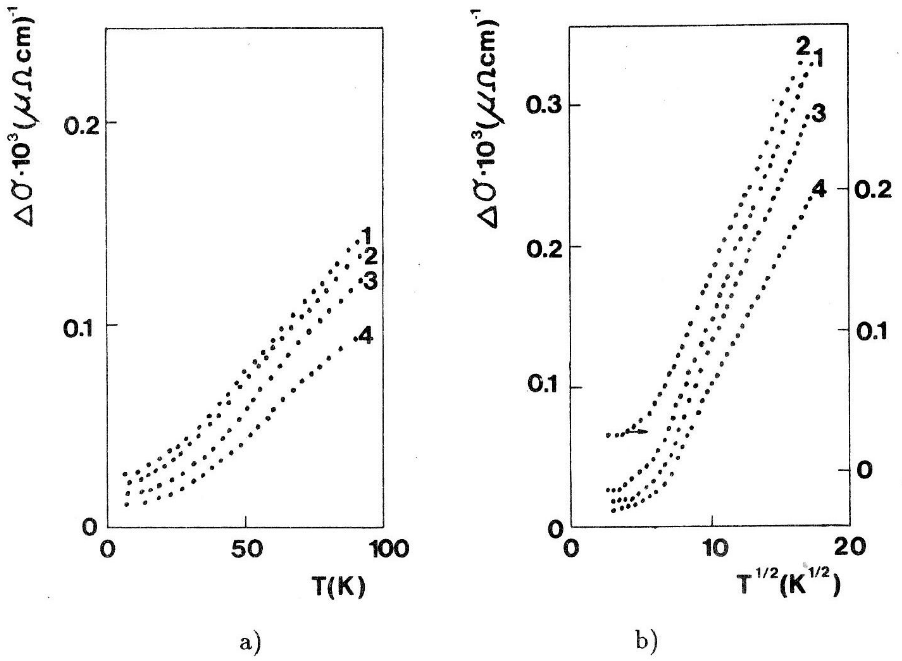

Fig. 1. Change in electric conductivity $\Delta\sigma = \sigma_T - \sigma_0$ for selected $Zr_2(Ni_{1-x}M_x)_1$ *glassy alloys vs. T (a) and $T^{1/2}$ (b): (1)* $Zr_2(Ni_{0.9}Co_{0.1})_1$, *(2)* $Zr_2(Ni_{0.8}Ti_{0.2})_1$, *(3)* $Zr_2(Ni_{0.5}Fe_{0.5})_1$ *and (4)* $Zr_2(Ni_{0.9}Mn_{0.1})_1$.

deviations from linearity [2] observed both at the lowest and highest temperatures in Fig. 1a, Figs. 1a and b taken together support the analysis of $\Delta\sigma$ in terms of the dominant T and $T^{1/2}$ contributions at lower and higher temperatures, respectively. Therefore, as for the binary Zr-3d alloys $\Delta\sigma$ of the ternary $Zr_2(Ni_{1-x}M_x)_1$ alloys seems to be dominated by the effects of the incipient localisation. From the experimental data for $\Delta\sigma$ we have determined the values of the coefficients A and B associated with the low temperature T and high temperature $T^{1/2}$ variation of $\Delta\sigma$, respectively. As explained before, these coefficients are related to the strength of the electron-phonon coupling in the investigated alloys. The values of A and B are given in Table 1 together with the other relevant data for $Zr_2(Ni_{1-x}M_x)_1$ alloys.

In what follows we shall proceed with the discussion of the data collected in Table 1. We will also compare these results with those obtained from the heat capacity measurements [10,11] and the photoemission experiments performed on the similar ternary $(Zr_{67}Ni_{33})_{85}M_{15}$ alloys [10].

As seen from Table 1, the resistivities ρ at 273 K of all $Zr_2(Ni_{1-x}M_x)_1$ alloys deviate only a little from that of Zr_2Ni. We note that a weak (incipient) localisation is expected to have a minor influence [2] on the residual conductivity σ_0. Because of this the magnitude of the resistivity is expected to be adequately described by some kind of the classical Boltzmann type theory (such as the Ziman or the

Mott model) [9,12]. Accordingly, since a rather small amount (x) of M is not expected to change much either structural or electronic parameters of amorphous Zr_2Ni matrix, any larger variation of ρ_{273} with M appears to be unlikely. We note, however, that the values of ρ_{273} for M to the left of Fe in the 3d transitions series are systematically somewhat lower than those for the elements to the right of Fe. This result combined with the evidence provided from the photoemission and UPS experiments (which will be discussed below) may indicate some contribution of d-electrons to the residual conductivity of amorphous $Zr_2(Ni_{1-x}M_x)_1$ alloys [13]. However, considering the fact that the observed variation of ρ_{273} with M is just outside of the error of the absolute magnitude of ρ_{273}, any further discussion of this point would be premature.

The temperature coefficients of resistivity at the room temperature $\alpha_{273} = \rho_{273}^{-1}(\mathrm{d}\rho/\mathrm{d}T)_{273}$ are also listed in Table 1. The values of α_{273} for $Zr_2(Ni_{1-x}M_x)_1$ alloys show considerably more variation than the corresponding ρ_{273} values. This and the observed $T^{1/2}$ variation of $\Delta\sigma$ above 100 K are difficult to understand within the framework of the classical theory such as the Faber-Ziman one and support our former conclusion that $\Delta\sigma$ of these alloys is dominated by the contributions arising from the incipient localisation. Because of this we will discuss the temperature dependence of resistivity of $Zr_2(Ni_{1-x}M_x)_1$ alloys in terms of the coefficients A and B rather than α_{273}.

Next we discuss the properties which are more directly related to the electronic structure of our alloys. The room temperature magnetic susceptibilities χ_p of $Zr_2(Ni_{1-x}M_x)_1$ alloys, which are also listed in Table 1, show an increase in going from M = Cu towards M = Ti. In addition to this general tendency, the magnetic susceptibilities of $Zr_2(Ni_{1-x}M_x)_1$ alloys with M close to the middle of 3d series seem to be enhanced. These features are more clearly seen from the excess susceptibility $\Delta\chi$, defined as the difference between the susceptibility of the particular $Zr_2(Ni_{1-x}M_x)_1$ alloy and Zr_2Ni alloy per mole of M [4]. Figure 2 shows that $\Delta\chi$ increases monotonically from M = Cu towards M = Ti and in addition to that exhibits a sharp maximum centered at M = Mn. Such an enhancement of $\Delta\chi$ for M around the middle of 3d-series is well known from the investigations of the dilute alloys of normal metals with 3d-impurities and is linked to the tendency of the formation of localized magnetic moment at the impurity site [14]. Therefore, we tentatively ascribe the maximum of $\Delta\chi$ centered at M = Mn in our alloys either to the formation of local magnetic moment at the M site or to the enhancement of the localized spin fluctuations in this range of M (if the stable local moment does not form). As shown below, this assumption is supported by the results of the heat capacity and superconducting transition temperature measurements. At the same time the background monotonic increase of $\Delta\chi$ on moving from Cu towards Ti should be linked to an increase of the electronic density of states (EDOS) at the Fermi level $g(E_F)$ caused by the addition of M to Zr_2Ni matrix.

The above suggestion is supported by the EDOS of the similar amorphous $(Zr_{67}Ni_{33})_{85}M_{15}$ alloys obtained from UPS He I ($h\nu = 21.2$ eV) experiments [10]. EDOS of amorphous Zr_2Ni matrix shows two maxima, one at E_F ($E = 0$) belonging to Zr 4d-states and the other centered around $E = -1.8$ eV belonging to

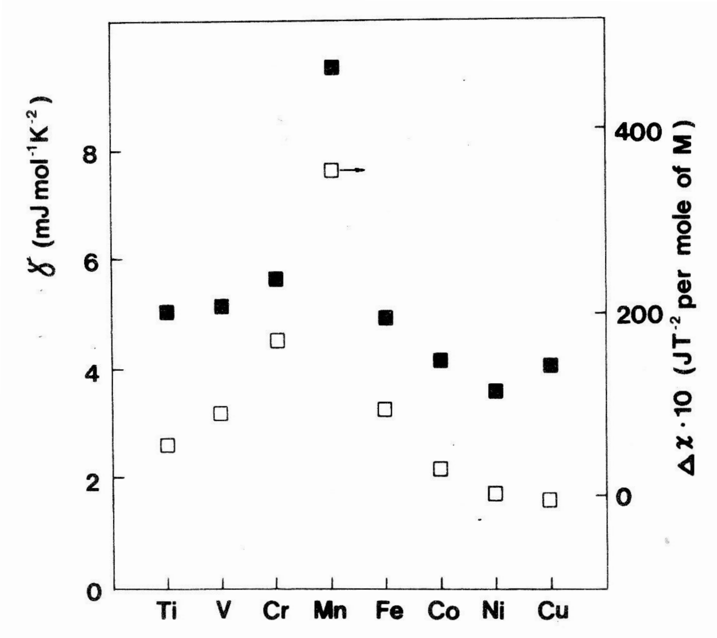

Fig. 2. Electronic specific heat coefficient γ for amorphous $(Ni_{33}Zr_{67})_{85}M_{15}$ alloys (■) and the change in magnetic susceptibility $\Delta\chi$ (□) per mole of M of amorphous $Zr_2(Ni_{1-x}M_x)_1$ alloys vs M.

Ni 3d-states. The replacement of Ni with M = Ti enhances the maximum at E_F (therefore enhences $g(E_F)$) and reduces the other one, whereas the replacement of Ni by Cu reduces both maxima (therefore reduces $g(E_F)$). The behaviour of EDOS at E_F for M between Ti and Cu follows the general tendency observed in $\Delta\chi$ (Fig. 2) i.e. shows that $g(E_F)$ increases on going from M = Cu towards M = Ti.

A more quantitative insight into the variation of the EDOS at E_F with M can be obtained from the heat capacity measurements at low temperatures performed on the same alloy system [10]. The coefficients γ of the linear term in the low temperature heat capacity of amorphous $(Zr_{67}Ni_{33})_{85}M_{15}$ alloys are shown in Fig. 2 together with our data for $\Delta\chi$ of $Zr_2(Ni_{1-x}M_x)_1$ alloys. The variation of γ with M is almost identical to that of $\Delta\chi$. γ increases in going from M = Cu towards M = Ti and in addition shows an enhancement for M = V, Cr, Mn and Fe exhibiting a maximum at M = Mn. It is well known [14] that the magnetic effects (such as the local moment formation or the localized spin fluctuations) enhance γ in a similar way as χ. Therefore the results for γ confirm the conclusions reached from our analysis of $\Delta\chi$ and also enable one to determine $g(E_F)$, providing that the strength of the electron-phonon coupling is known.

Figure 3 shows the variation of the coefficients A and B (associated with the approximate T and $T^{1/2}$ terms in $\Delta\sigma$, respectively) of $Zr_2(Ni_{1-x}M_x)_1$ alloys with M. As discussed earlier in this paper these coefficients are related to the strength of the

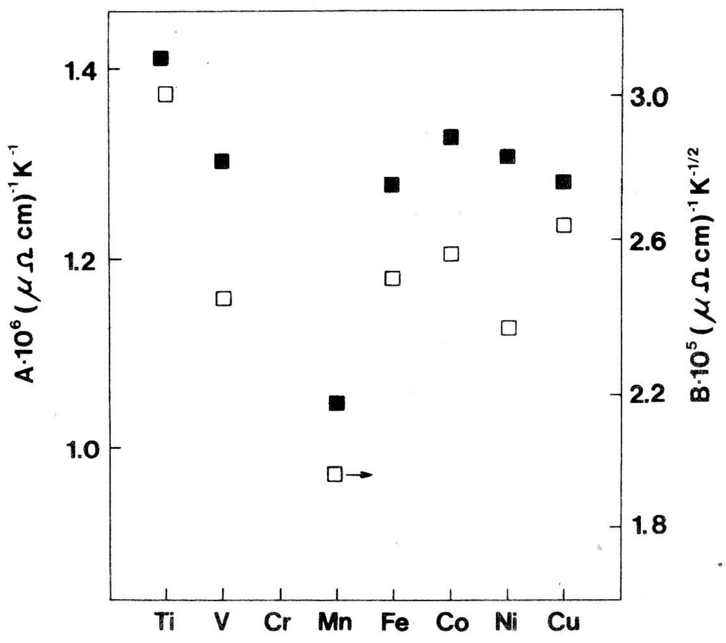

Fig. 3. Coefficient A of linear conductivity variation bellow 100 K *(■) and coefficient B of a $T^{1/2}$ conductivity variation (□) above 100* K *vs* M.

electron-phonon coupling via τ_i but due to the neglect of τ_{s0} and of other contributions [2] affecting $\Delta\sigma$ at lower temperatures this relation is at the best qualitative (for a more quantitative determination of this relationship the measurements of the magnetoresistance [8] at low temperatures are required). Within the experimental error in the determination of A and B (which is quite large because the resistance changes only a little with temperature in amorphous alloys) both A and B exhibit a variation with M quite similar to that observed for γ of $(\mathrm{Zr_{67}Ni_{33}})_{85}\mathrm{M}_{15}$ alloys (Fig. 2). In particular A and B tend to increase on going from M = Cu towards M = Ti and exhibit a sharp minimum at M = Mn. This similarity seems to support our suggestions that the variation of $\Delta\sigma$ in amorphous $\mathrm{Zr_2(Ni_{1-x}M_x)_1}$ alloys is dominated by the incipient localisation effects whose strength depends on the electron-phonon coupling. The minimum of A and B centered at M = Mn is apparently connected with the magnetic interactions and is consistent with the theoretical predictions for the incipient localisation [5].

Within the framework of our explanation of the variation of $\Delta\sigma$ in $\mathrm{Zr_2(Ni_{1-x}M_x)_1}$ alloys the depression of A and B around M = Mn can be described in terms of the reduction of the effective electron-phonon coupling. Such an effective reduction in the electron-phonon coupling is consistent with the variations of the superconducting transition temperatures T_c in $\mathrm{Zr_2(Ni_{1-x}M_x)_1}$ alloys. In particular, the gradients of the change in T_c, $\Delta T_c/\Delta x$ [4] (which could be by means of the McMillan expression [15] related to the electron-phonon coupling constant) showed quantitatively very similar variation with M to those of A and B. However, in order to perform a more quantitative analysis of the superconducting transitions temperatures [16] the measurements of the low temperature heat capacities on the same samples are required.

4. Conclusion

The change of conductivity $\Delta\sigma$ of the ternary amorphous $Zr_2(Ni_{1-x}M_x)_1$ alloys varies with temperature in qualitatively the same way as that of the amorphous Zr-3d alloys (3d = Fe, Co, Ni and Cu) and therefore has the same physical origin. The observed variations $\Delta\sigma \sim T$ at lower and $T^{1/2}$ at higher ($T > 100$ K) temperatures are reminiscent of the incipient localisation which is quite likely to occur in these high-resistivity alloys. Like in Zr-3d alloys [1,2] the other contributions to $\Delta\sigma$ (both classical and quantum) affect the results at the lowest temperatures ($T < 20$ K).

The importance of these alloys is, however, that the addition of M to Zr_2Ni matrix introduces some novel features in both the electronic transport and the thermodynamic properties which have not been observed in the Zr-3d alloys containing late transitions metals only. In particular, in Zr-3d alloys all the properties varied rather slowly and monotonically with the 3d content as if the effect of 3d element was merely the dilution of amorphous Zr. This was the consequence of the specific electronic band structures of the alloys of Zr with the late 3d-elements in which only the Zr-4d states were present at the Fermi level whereas the 3d-states were at considerably higher binding energies.

In $Zr_2(Ni_{1-x}M_x)_1$ alloys (where M can be any member of 3d series) already small amount (x) of M can produce considerable change in the thermodynamic and the electronic transport properties. The reason for this is twofold: the modifications in the electronic band structures of the alloy [10] as M changes from Cu towards Ti and the magnetic state of M. The tendency for the formation of the localized magnetic moments for M around the middle of 3d series has particularly strong effects on the properties of investigated alloys. Whereas the effects of local moment formation (or spin fluctuations) on the thermodynamic and superconducting properties of amorphous $Zr_2(Ni_{1-x}M_x)_1$ alloys are qualitatively the same as those observed in dilute (crystalline) alloys of normal and noble metals with 3d-impurities [14], their effects on the electronic conduction are quite different in two cases.

This difference probably arises from the competition between the incipient localisation and magnetic interactions in $Zr_2(Ni_{1-x}M_x)_1$ alloys. The measurements of the magnetoresistance and the low temperature heat capacities on these alloys should enable a more quantitative analysis of the reported phenomena and may also bring a new insight into the competition of the incipient electron localisation and magnetism.

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ELEKTRONSKA I MAGNETSKA SVOJSTVA TERNARNIH $Zr_2(Ni_{1-x}M_x)_1$ STAKLASTIH SLITINA

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Mjereni su električni otpori (u temperaturnom intervalu između 8 K i 300 K), magnetska susceptibilnost na sobnoj temperaturi i temperatura supravodljivog prijelaza za amorfne slitine $Zr_2(Ni_{1-x}M_x)_1$ (M = Ti, V, Cr, Mn, Fe, Co, Ni i Cu). Kao i za druge Zr-3d staklaste slitine (3d = Fe, Co, Cu ili Ni) temperaturna ovisnost električnog otpora može se objasniti efektom početne lokalizacije koji je svojstven slitinama s visokim električnim otporom. Nova svojstva nastala uvođenjem M pojava su nejednolikih promjena magnetskih i električnih svojstava s M. Uzrok su tih promjena sustavne promjene u elektronskoj strukturi kada idemo od M = Cu do M = Ti (to je potvrđeno eksperimentima fotoemisije na sličnim $(Zr_{67}Ni_{33})_{85}M_{15}$ amorfnim slitinama) i sklonost prema formiranju lokaliziranih magnetskih momenata za M oko sredine 3d-serije (M = V, Cr, Mn i Fe). Neobično je svojstvo dosta jako prigušenje utjecaja efekata početne lokalizacije na električni otpor uslijed magnetskih interakcija.