

# Magnetic and Transport properties of Co-Cu Microwires

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**Abstract**—We report on the magnetic, transport and structural properties of  $\text{Co}_x\text{-Cu}_{100-x}$  ( $5 \leq x \leq 40$ ) glass-coated microwires. For  $x=5$  we observed the resistivity minimum at 40 K associated with the Kondo effect. For  $x \geq 10$  we observed considerable magnetoresistance effect. Temperature dependence of susceptibility show considerable difference for  $x > 10$  and  $x \leq 10$  attributed to the presence of small Co grains embedded in the Cu matrix for  $x \geq 10$ . Using X-ray diffraction we found, that the structure of  $\text{Co}_x\text{-Cu}_{100-x}$  microwires  $x \geq 10$  is granular consisting of two phases: fcc Cu appearing in all the samples and fcc  $\alpha$ -Co presented only in microwires with higher Co content.

**Keywords:** magnetic microwires; GMR effect; internal stresses; Kondo effect

## I. INTRODUCTION

Studies of various materials containing small nano-sized magnetic particles, grains or even magnetic impurities in non-magnetic matrix attracted great attention owing to a number of interesting properties, such as giant magnetoresistance (GMR) effect or Kondo effect [1-6]. Granular materials consisting of small grains distributed inside a non-magnetic matrix exhibiting giant magnetoresistance (GMR) effect have attracted considerable attention since beginning of 1990-s [1,3]. Mostly granular materials have been obtained in systems of immiscible elements (like Co-Co, Co-Ag...), when the granular structure can be obtained by the precipitation of small ferromagnetic grains from the supersaturated solid solutions [8]. Like in the case of multilayered thin films, GMR effect has been attributed to spin-dependent scattering of conduction electrons within the magnetic granules as well as at the interfaces between magnetic and nonmagnetic regions [1,3,7]. The size and the spatial distribution of nano-sized ferromagnetic grains (usually of Co or Fe) inside the metallic matrix (typically Cu, Ag, Au or Pt) are crucial for obtaining high GMR effect. On the other hand, if the size magnetic grains is small and even if they can be considered as magnetic impurities, the scattering of conduction electrons in a metal due to magnetic impurities give rise to a Kondo effect [2,5,6,8].

Development of novel materials (like graphene) allows extending the systems where Kondo effect can be realized [8]. On the other hand, Kondo effect has become a key concept in

condensed matter physics in understanding the behavior of metallic systems with strongly interacting electrons. Development of nano-scaled electronic devices requires the understanding and the control of electron behavior, in particular, of correlation effects at the atomic scale. The Kondo effect related to the resonant scattering of conductive electrons by quantum local centers is one of the key correlation effects in condensed matter physics.

As mentioned above in the case of immiscible alloys the structure consisting of nano-sized ferromagnetic grains randomly distributed in metallic matrix can be realized by the recrystallization of metastable alloys produced by various techniques. One of the common methods for obtaining such granular materials is a melt spinning technique involving rapid quenching from the melt [9-11].

Following general trend on miniaturization of modern devices and sensors the studies of composite thin wires consisted of metallic nucleus surrounded by glass coating gained considerable attention during last few years. Such microwires are produced by so-called Taylor-Ulitovski technique [10-12] allowing fabrication of glass-coated metallic microwires (typical metallic nucleus diameters 1 - 30  $\mu\text{m}$ , the thickness of the glass coating 0,5 - 20  $\mu\text{m}$ ). The fabrication method denominated in most of modern publications as a modified Taylor-Ulitovsky and/or quenching-and-drawing method is well described in recent publications [10-14].

Main advantages of this technique are high quenching rate, homogeneous geometry of metallic nucleus and glass coating and almost continuous process allowing fabrication of km long wires. Consequently preparation of microwires with amorphous, nanocrystalline, microcrystalline or granular structure of metallic nucleus is reported [10-14].

Until now most interest is paid to studies of amorphous and nanocrystalline microwires excellent and unusual soft magnetic properties such as fast magnetization switching related with large Barkhausen jump [11,14] and the giant magnetoimpedance (GMI) effects [10-11,15]. Particularly glass-coated microwires exhibiting high GMI effect demonstrated extremely high magnetic field sensitivity (up to pT) and recently found industrial application in magnetic field sensors [15,16].

On the other hand recently reported on preparation of microwires exhibiting magnetocaloric, shape memory and GMR effects [10,11].

One of the peculiarities of the Taylor-Ulitovsky technique is the composite character of microwires and, therefore, the strong internal stresses induced during the simultaneous rapid solidification of metallic nucleus surrounded by the glass coating [11,15-19]. The structure and magnetic properties of glass-coated microwires are drastically affected by the existence of the outer glass-coating with physical properties completely different from metallic nucleus alloys (different thermal conductivity, thermal expansion coefficients...) [15,19,20]. For example, crystalline structure, crystallization temperature and magnetic properties of nanocrystalline microwires obtained by recrystallization of the amorphous precursor are rather different from nanocrystalline ribbons of the same composition [19,20]. The strength of internal stresses is determined by the  $\rho$ -ratio of metallic nucleus diameter,  $d$ , and total diameter,  $D$  ( $\rho=d/D$ ).

The effect of internal stresses on structure and properties of solids obtained using rapid quenching is related with the non-equilibrium thermodynamics. For discussion of the structure of the crystalline phases one should take into account the nucleation and crystalline growth process related with atomic diffusion under stress[21]. Consequently the strength of internal stresses might be tailored controlling the  $\rho$ -ratio [18,19]. Accordingly the Taylor-Ulitovsky method allows tuning the structure and the properties of the composite wires controlling the fabrication parameters and preparing the microwires with different  $\rho$ -ratios [19, 20]. We can consider that in the case of microwires consisting of immiscible elements the internal stresses can affect structure and properties.

In this work we report on our recent studies of  $\text{Co}_x\text{-Cu}_{100-x}$  (5 at % $\leq x \leq$  40 at %) glass-coated microwires and on observation of Kondo-like behaviour and GMR effect in Co-Cu microwires.

## II. EXPERIMENTAL DETAILS

We prepared a number of  $\text{Co}_x\text{-Cu}_{100-x}$  (5 $\leq x \leq$ 40 at %) glass-coated microwires (typical total diameters,  $D$ , from few to 30  $\mu\text{m}$ ) consisted of metallic nucleus (typical diameters,  $d$ , 9- 20  $\mu\text{m}$ ) covered by outer glass shell (typical thickness from 3  $\mu\text{m}$  till 10  $\mu\text{m}$ ) using the Taylor-Ulitovsky technique (see details in table 1) [10-15]. Within each composition of metallic nucleus we produced microwires with different ratio of metallic nucleus diameter,  $d$ , and total diameter,  $D$ , i.e. with different ratios  $\rho=d/D$ . This allowed us to control residual stresses, since the strength of internal stresses as described above is determined by the ratio  $\rho$  [18,19].

In the laboratory process, an ingot containing few grams of the master alloy with the desired composition is placed into a Pyrex-like glass tube and within a high frequency inductor heater. The alloy is heated up to its melting point, forming a droplet. While the metal melts, the portion of the glass tube adjacent to the melting metal softens, enveloping the metal droplet (Fig.1). A glass capillary is then drawn from the

Table 1. Compositions and geometric properties of studied  $\text{Co}_x\text{-Cu}_{100-x}$  microwires

Microwires composition	Microwires geometry: metallic nucleus diameter, $d$ , total diameter, $D$ and ratio $\rho=d/D$
$\text{Cu}_{95}\text{Co}_5$	$d=13,3 \mu\text{m}$ ; $D=16,9 \mu\text{m}$ , $\rho=0.787$
$\text{Cu}_{90}\text{Co}_{10}$	$d=11.6 \mu\text{m}$ ; $D=15.4 \mu\text{m}$ ; $\rho=0.753$ $d=14.7 \mu\text{m}$ ; $D=20.2 \mu\text{m}$ ; $\rho=0.728$
$\text{Cu}_{80}\text{Co}_{20}$	$d=13.3 \mu\text{m}$ ; $D=20.5 \mu\text{m}$ ; $\rho=0.649$ $d=14.4 \mu\text{m}$ ; $D=20.5 \mu\text{m}$ ; $\rho=0.702$ : $d=12.9 \mu\text{m}$ ; $D=18 \mu\text{m}$ ; $\rho=0.717$ :
$\text{Cu}_{70}\text{Co}_{30}$	$d=9\mu\text{m}$ ; $D=14.7 \mu\text{m}$ ; $\rho=0.612$ $d=11.2\mu\text{m}$ ; $D=16.9 \mu\text{m}$ , $\rho=0.663$ : $d=18.4\mu\text{m}$ ; $D=24.1 \mu\text{m}$ ; $\rho=0.763$
$\text{Cu}_{60}\text{Co}_{40}$	$d=15.4 \mu\text{m}$ ; $D=27.7 \mu\text{m}$ ; $\rho=0.57$ $d=9.4 \mu\text{m}$ ; $D=15.1 \mu\text{m}$ ; $\rho=0.62$

softened glass portion and wound on a rotating coil. At suitable drawing conditions, the molten metal fills the glass capillary and a microwire is thus formed where the metal core is completely coated by a glass shell.

Structure and phase composition have been checked using a X-ray diffractometer with  $\text{Cu K}_\alpha$  ( $\lambda=1.54 \text{ \AA}$ ) radiation (model BRUKER D8 Advance).

We used Physical Property Magnetic System (PPMS) device for measurements of magnetic and magneto-transport properties in the temperature range 5 - 300 K.

The magnetoresistance (MR) has been defined as:

$$\Delta R/R(\%) = (R(H)-R(0)) \times 100/R(0) \quad (1)$$

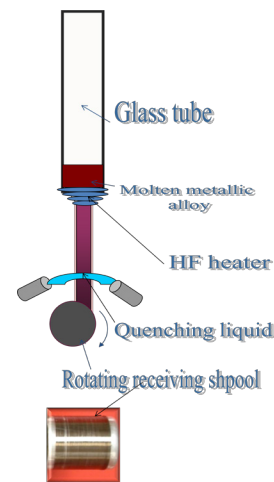


Figure 1. Schematic picture of the fabrication process allowing preparation of glass-coated microwires

### III. RESULTS AND DISCUSSION

For  $\text{Co}_x\text{-Cu}_{100-x}$  microwires at  $10 \leq x \leq 30$  we observed considerable MR effect (Fig2). In the case of lowest Co content ( $\text{Co}_5\text{Cu}_{95}$  microwires) the MR effect is much lower. Decreasing the temperature,  $T$ , the  $\Delta R/R$  increases. For  $\text{Co}_{20}\text{Cu}_{80}$  microwires at  $T=5\text{K}$  we observed  $\Delta R/R \approx 8\%$ .

We observed a well pronounced minimum on the temperature dependence of resistance,  $R$ , at about  $T_k \approx 40\text{K}$  (Fig.3), which should be attributed to Kondo-like behaviour. Few different mechanisms might be considered to explain an

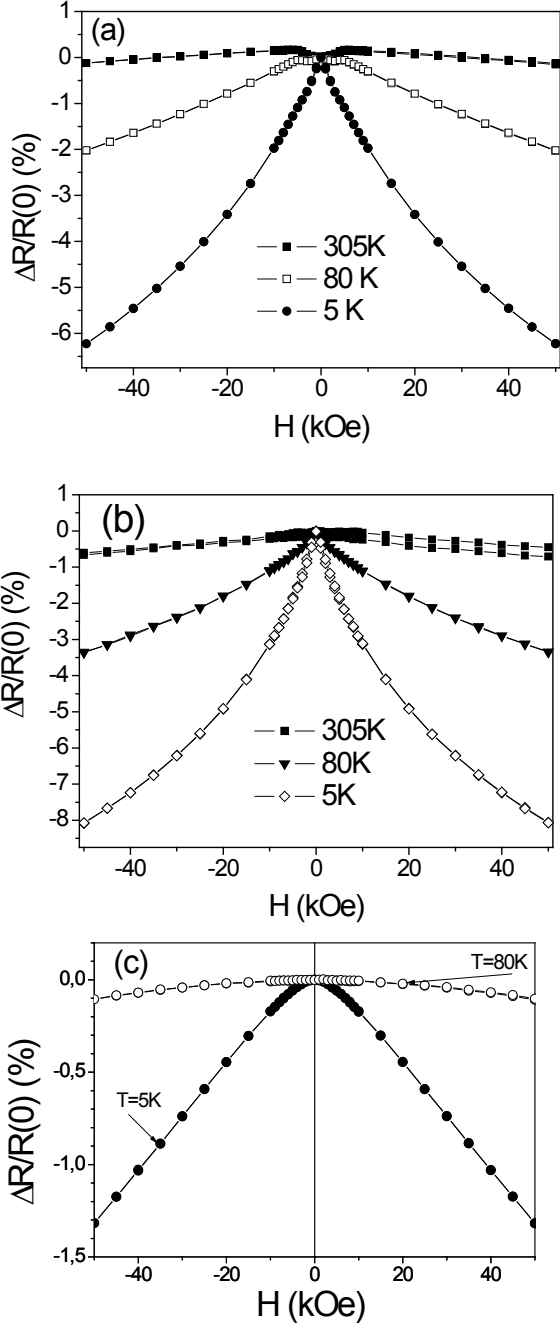


Figure 2.  $\Delta R/R(H)$  dependences for  $\text{Co}_x\text{Cu}_{100-x}$  microwires with  $x=30$ ,  $\rho=0.612$  (a) and  $x=20$ ,  $\rho=0.649$  (b) and with  $x=5$ ,  $\rho=0.787$  (c) measured at different temperatures with maximum field of 50 kOe .

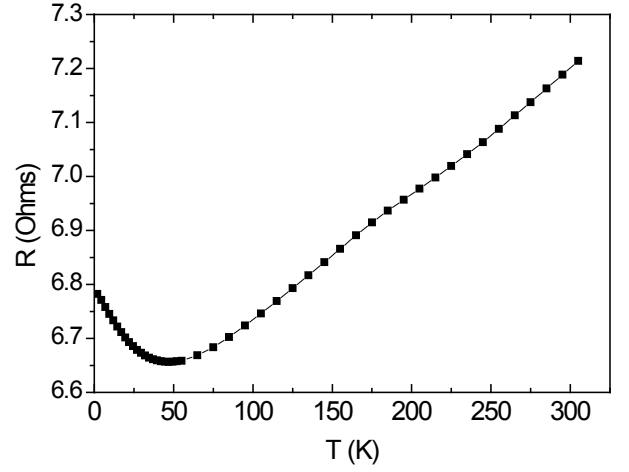


Figure 3. Temperature dependences of resistivity  $\text{Co}_5\text{Cu}_{95}$  sample

increase of the resistivity of a metallic microwire at low temperatures: magnetic Kondo effect, weak localization, enhanced electron-electron interaction, scattering of conduction electrons by structural two-level system (TLS) and scattering of strongly spin-polarized charge carriers on diluted magnetic moments [21-24]. Figure 3 shows that in the present case the temperature dependence of the resistivity below 40 K has a form typical for the Hamann model [25]. Furthermore, magnetic field sufficiently affects the logarithmic growth of resistivity making it much weaker (Fig.4).

MR is negative in this temperature range that also corresponds to suppression of Kondo effect in strong magnetic fields. On the other hand the resistivity minimum is not completely suppressed even at maximum magnetic field,  $H=7\text{ T}$ , the temperature of resistivity minimum is rather high for the diluted Kondo alloys, the concentration of randomly distributed Co ions in Cu matrix is not low (that can lead to interaction between magnetic ions and, consequently to partial suppression of Kondo effect). Therefore we cannot exclude the role of TLS mechanism.

Consequently, we assume that the most appropriate explanation of observed minimum on the temperature dependence of resistance,  $R$ , at about 40K is the Kondo effect and/or TLS.

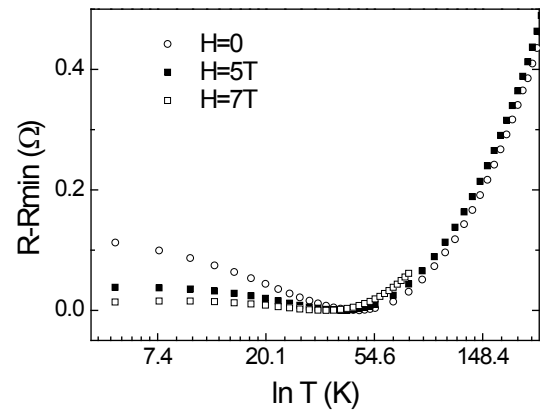


Figure 4. Effect of magnetic field on temperature dependence of resistivity,  $R$ , of  $\text{Co}_5\text{Cu}_{95}$  microwires.

In fact,  $\text{Co}_5\text{Cu}_{95}$  alloy is not a classical diluted Kondo system not only because of large Co single-ion concentration but also because a part of Co ions forms magnetic clusters and these clusters will create local magnetic fields and spin-polarization of current carriers [25]. The presence of such clusters can be considered from the magnetization,  $M$ , measurements at different temperatures. Particularly we observed magnetization saturation at low temperatures (Fig.5).

Fig. 6 shows that in contrast to the samples with higher content of Co,  $\text{Co}_5\text{Cu}_{95}$  and  $\text{Co}_{10}\text{Cu}_{90}$  microwires do not present blocking temperature down to 5 K and do not present non vanishing susceptibility,  $\chi$ , at higher temperature typical

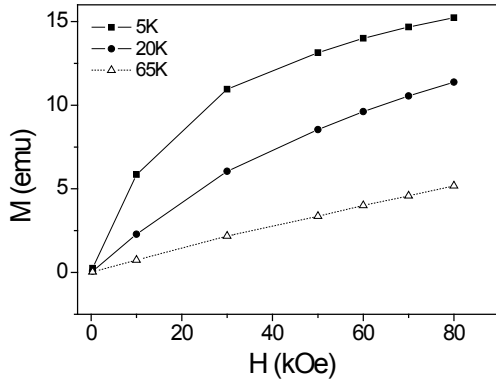


Fig.5 Magnetization curves of  $\text{Co}_5\text{Cu}_{95}$  microwires measured at different temperatures.

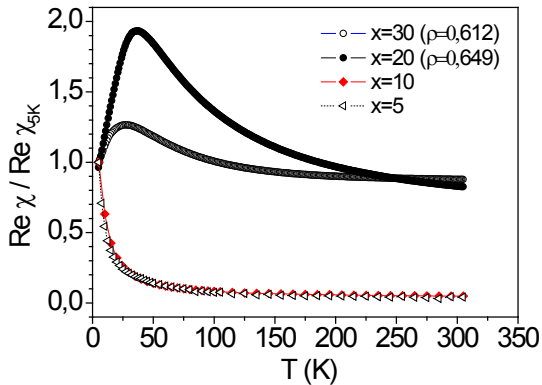


Figure 6. AC-susceptibility measured for various  $\text{Co}_x\text{Cu}_{100-x}$  microwires

for multi-domain cobalt clusters. Apparently this interaction is strong enough in the  $\text{Co}_{10}\text{Cu}_{90}$  sample to allow significant suppression of magnetic disorder at moderate fields giving rise to considerable negative magnetoresistance (see Fig. 2). Co clusters must be small enough to have a blocking temperature below 5 K, but their interaction is appreciable and affects  $M$  vs  $H$  curves at low temperatures.

Interaction between the clusters in the  $\text{Co}_5\text{Cu}_{95}$  sample is weaker, that results in very low magnetoresistance. Pronounced Kondo-type minima in resistivity shows that considerable part of Co atoms is not magnetically ordered and therefore not affected by the local fields of clusters.

At the same time the local fields of the clusters seem to be quite strong and cause the ordering of the moments of nearby

Co atoms suppressing Kondo-type scattering (sample with  $x=10$  does not exhibit minimum on temperature dependence of resistivity).

In order to analyse the origin of GMR effect we compared temperature dependence of  $\Delta R/R$  and  $M^2$  for  $\text{Cu}_{90}\text{Co}_{10}$  with  $\rho=0.728$  and for  $\text{Cu}_{80}\text{Co}_{20}$  microwire with  $\rho\approx 0.7$  (see Fig.7). We observed qualitative correlation for all temperature range typical for granular systems.

X-ray diffraction (XRD) has been employed for structural studies. For  $x\geq 10$  the structure of the metallic core is granular and consists of two phases: the main phase - *fcc* Cu (lattice parameter 3.61 Å), found in all samples and *fcc*  $\alpha$ -Co (lattice

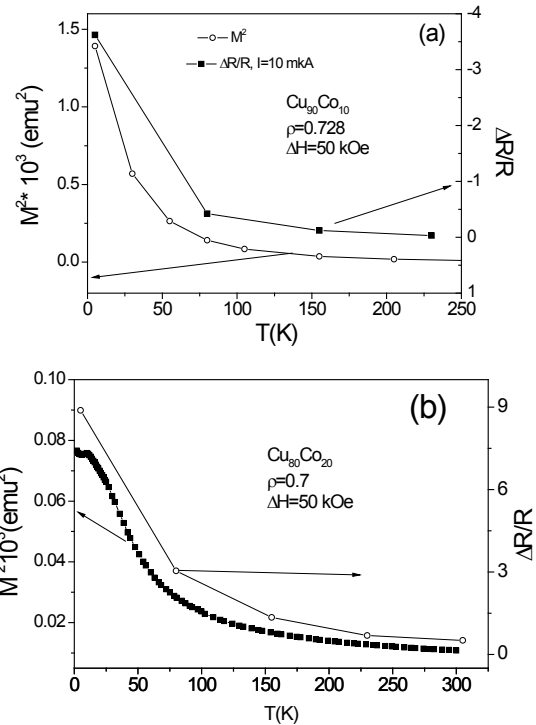


Figure 7. Temperature dependences of  $\Delta R/R$  and  $M^2$  for  $\text{Cu}_{90}\text{Co}_{10}$  and  $\text{Cu}_{80}\text{Co}_{20}$  microwires

parameter 3.54 Å) which presents in microwires with higher Co content. In the  $\text{Co}_5\text{Cu}_{95}$  sample we observed only *fcc* Cu: Co atoms are distributed within the Cu crystals (Fig.8a).

The quantity and the crystallite size of the formed phases strongly depend on the geometry of the microwires ( $\rho$ -ratio). The residual concentration of Co dissolved in the Cu matrix and size of Co grains depend on the quenching rate and on internal stresses determined by the  $\rho$ -ratio. Consequently, strong internal stresses induced during the simultaneous rapid solidification of the thin metallic wire and glass coating layer affect magnetic properties and structure of glass coated microwires.

Fig.8b shows XRD for  $\text{Co}_{40}\text{Cu}_{60}$  and  $\text{Co}_{10}\text{Cu}_{90}$  microwires. We can see that in microwires with  $x=40$   $\alpha$ -Co phase (average grain size about 40 nm) has been observed. At the same time we did not observe this phase in the sample with  $x=10$ . It means that in  $\text{Co}_x\text{-Cu}_{100-x}$  microwires for  $x\leq 10$  the room

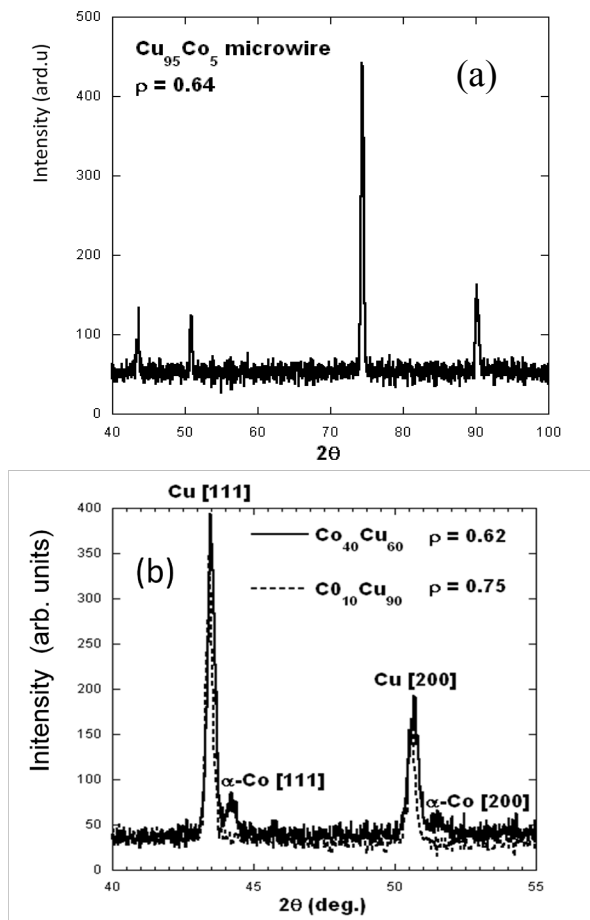


Figure 8. XRD of  $\text{Co}_5\text{Cu}_{95}$  microwire ( $\rho=0.64$ ) measured at room temperatures (a) and  $\text{Co}_{10}\text{Cu}_{90}$  and  $\text{Co}_{40}\text{Cu}_{60}$  microwire microwires ( $\rho=0.75$  and  $0.62$  respectively).

temperature structure of the samples is the supersaturated solid solution of Co atoms or small clusters in Cu matrix.

This is consistent with the Co-Cu equilibrium phase diagram and the method of microwires fabrication involving rapid quenching from the melt. Indeed, the Co-Cu phase diagram exhibit at room temperature almost immiscibility, although at elevated temperature there is solubility of Co in Cu of about 13 at%. Therefore we can assume that rapid quenching results in quenching of high temperature structure consisting in supersaturated Co solution in Cu matrix for  $x \leq 13\%$ .

Additionally, our data can be interpreted considering, that for  $\text{Co}_5\text{Cu}_{95}$  sample Co atoms are distributed between the matrix and very small clusters (Fig.8). For concentrations  $x > 10$  partial coalescence of granules was observed. The anisotropic contribution to MR has been observed for  $x \geq 30$ , giving rise to non-monotonic magnetic field dependence of MR (Fig2b).

It is worth mentioning, that previously Kondo-like behaviour in Co-Cu system has been observed only in clusters consisting of a single Co atom and several Cu atoms fabricated by sophisticated technique using atomic manipulation with the microscope tip [26]. On the other hand previously resistivity minimum versus temperature has been

observed in Co-Ag thin films prepared by electron gun evaporation under ultrahigh-vacuum conditions [27]. Like in our case, weak-localization effects have not been considered for interpretation of the observed resistivity minima considering low absolute values of the residual resistivity [27]. An interpretation associated with loose spins proposed by Slonczewski [28] and the decreasing of the spin-dependent scattering responsible for the GMR overwhelming an increasing spin fluctuation scattering have been also discussed, although further theoretical analysis is suggested for the case of the granular systems. On the other hand, the logarithmic temperature dependence of resistivity has been observed for the case of Au/Fe superlattices [5]. The authors considered two types of loose spins (within the non-magnetic spacer layer the “loose interfacial spins” comprising the ferromagnetic layers). But since magnetic field strongly affects the logarithmic temperature dependence of resistivity, Kondo-like scattering is considered for the interpretation of results. In the latter case the logarithmic temperature dependence of resistivity has been considered as an intrinsic of the multilayer samples, since similar dependence has been observed by the same authors in Al/Ni and Al/Ag multilayers with low thicknesses of layers (below 5 nm) [29].

On the other hand, quite recently for interpretation of temperature dependence of resistivity of nano-scale granular Aluminum films, with Al grains weakly coupled through thin Al oxide barriers it was suggested that small metallic grains weakly coupled to their environment can behave like magnetic impurities in a metallic matrix [30]. It is also pointed out, that the weak electron localization also results in a negative magneto-resistance if spin-orbit scattering is negligible.

Low interaction between the magnetic particles is proved by good correlation  $\Delta R/R$  and  $M^2$  for  $\text{Cu}_{90}\text{Co}_{10}$  presented in Fig.7a. Usually deviations from  $\Delta R/R$  and  $M^2$  correlation are interpreted as the existence of interaction among the magnetic particles [9,31]. Some deviations from good correlation  $\Delta R/R$  and  $M^2$  observed for  $\text{Cu}_{80}\text{Co}_{20}$  microwire might be related with more considerable interaction between the magnetic particles for this microwire with higher Co content.

Fabrication method of microwires involves rapid quenching from the melt of the composite microwire. The existence of the outer glass-coating with different thermal expansion coefficient drastically affects the structure and the magnetic properties of glass-coated microwires. Therefore the microstructure of studied  $\text{Co}_5\text{Cu}_{95}$  microwires is quite different from that of thin films and multilayers studied before. The similarity might be originated by the existence of ultra-small Co grains and clusters distributed within the Cu crystals, as observed by the X-ray diffraction (Fig.8) and recently reported by us [32]. In this case similar results, as a pronounced minimum on the temperature dependence of resistance and considerable effect of magnetic field might be interpreted in similar way.

Consequently, although usually Kondo effect is considered for a magnetically diluted system we observed Kondo-like behavior in  $\text{Cu}_{95}\text{Co}_5$  microwires where Co atoms distributed between the matrix and very small clusters. It is worth mentioning, that we report on rather simple and fast (few



meters per minute) preparation of the Co-Cu microwire, where local structure can be manipulated through the control of the internal stresses.

#### IV. CONCLUSIONS

We observed GMR and Kondo-like behaviour in  $\text{Co}_x\text{Cu}_{100-x}$  microwires prepared by the Taylor-Ulitovsky method. For  $x=5\%$ , we observed resistivity minimum at 40 K. Magnetic field significantly affects temperature dependence of resistivity in  $\text{Co}_5\text{Cu}_{95}$  microwires. Additionally in  $\text{Co}_5\text{Cu}_{95}$  microwires by XRD we observed, that Co is distributed in the Cu- matrix. For  $x > 10$  evidences of granular structure have been observed and GMR effect is considerable higher. Temperature dependence of the AC-susceptibility show considerable difference for  $x \geq 20\%$  at low temperatures, related with the presence of small Co grains embedded in the Cu matrix. Anisotropic MR is observed in  $\text{Co}_x\text{Cu}_{100-x}$  microwires for  $x \geq 30$ . It is demonstrated that the local structure and magneto-transport properties can be manipulated through the control of the internal stresses. We propose rather simple and fast preparation method of Co-Cu microwire, where local structure can be manipulated through the control of the internal stresses.

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