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Negative magnetoresistivity of the RM_4Al_8 (R = Sc, Y, Ce, Yb, Lu; M = Cr, Mn, Fe) ternaries with the $ThMn_{12}$-type crystal structure

V.M. Dmitriev [a,b], A.V. Terekhov [a], W. Suski [b,c,*], L.A. Ishchenko [a], J. Ćwik [b],
T. Palewski [b], B.Ya. Kotur [d], E. Talik [e]

[a] *B. Verkin Institute for Low Temperature Physics and Engineering of the National Academy of Sciences of Ukraine, 47 Lenin Ave, 61103 Kharkov, Ukraine*
[b] *International Laboratory of High Magnetic Fields and Low Temperatures, P.O. Box 4714, 50-985 Wrocław 47, Poland*
[c] *W. Trzebiatowski Institute of Low Temperature and Structure Research, Polish Academy of Sciences, P.O. Box 1410, 50-950 Wrocław 2, Poland*
[d] *Department of Inorganic Chemistry, Ivan Franko National University of L'viv, vul. Kyryla i Mefodiya 6, 79005 L'viv, Ukraine*
[e] *A. Chełkowski Institute of Physics, Silesian University, 40-007 Katowice, Poland*

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Abstract

The phenomenon of negative magnetoresistivity (NM) at low magnetic fields has been detected in the RM_4Al_8 ternaries where R = Sc, Y, Ce, Yb, Lu, and M = Cr, Mn and Fe. The mechanisms responsible for this phenomenon have been carefully analyzed. It has been shown for the first time that the appearance of the NM for these compounds results from the Kondo effect combined with spin-glass state following crystallographic disorder.

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Keywords: Rare earth intermetallics; Negative magnetoresistance; Kondo effect

1. Introduction

During the last decades the phenomenon of the negative magnetoresistivity (NM)—a decrease of the electrical resistivity in the magnetic fields observed in various materials, has been intensively examined. The most popular and most frequently investigated materials are: the classical ferromagnets [1,2], granular structures of the nonmagnetic matrixes with the ferromagnetic impurities [3,4], the magnetic superstructures [5,6], and also manganites examined extensively recently [7,8]. Lately, the authors of Refs. [9–11] have discovered the new class of materials with the NM—the ternaries of the RM_4Al_8-type (where R—nonmagnetic lanthanide element or also Y and Sc, M—transition metal). These compounds exhibit the tetragonal body centered structure of the $ThMn_{12}$-type presented in Fig. 1 see (e.g. Ref. [14]). In this structure (space group: *I*4/*mmm*) the position 2(a) is occupied by the f-electron metals (lanthanides or actinides) as well as Y and Sc, the site 8(f) by the transition metal, whereas the sites 8(i) and 8(j) by the Al atoms.

In the present paper we try to answer the following questions:

(1) What is the influence of the change of the R element, when the M element is kept constant or reverse, on the magnitude and the temperature range of existence of the NM?
(2) What mechanisms could be responsible for the phenomenon of the NM in these compounds?

To get the answers to these questions the resistive and magnetoresistive properties of poly- and single crystalline samples with different R and M elements have been investigated in broad temperature (4.2–300 K) and magnetic field (0–0.7 T) ranges.

To shed light on the nature of the NM, below we discuss our experimental results in relation to the previously obtained data concerning the magnetic and structural characteristics, Kondo effect and spin-glass state of RM_4Al_8-type compounds.

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* Corresponding author at: W. Trzebiatowski Institute of Low Temperature and Structure Research, Polish Academy of Sciences, P.O. Box 1410, 50-950 Wrocław 2, Poland. Tel.: +48 71 343 50 21; fax: +48 71 344 1029.
E-mail address: w.suski@int.pan.wroc.pl (W. Suski).

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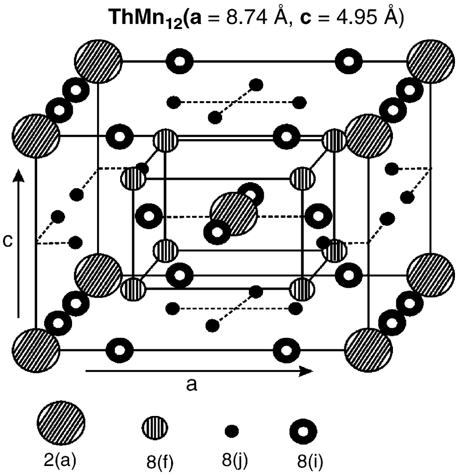

Fig. 1. Crystallographic unit cell of the $ThMn_{12}$-type structure. In the case of RM_4Al_8 compounds the R atoms are located at the 2(a); M—at the 8(f); Al—at the 8(i) and 8(j) sites. Lattice parameters of the $ThMn_{12}$ compound see e.g. Ref. [14].

2. Samples and measurements

In present work the following materials have been investigated: polycrystalline samples of $CeFe_4Al_8$, $ScFe_4Al_8$, $LuFe_4Al_8$, YCr_4Al_8 and YMn_4Al_8 obtained by arc- or radio-frequency melting, and single crystals of $ScFe_4Al_8$ and YFe_4Al_8 grown by Czochralski method—pulled out from the stoichiometric melt of initial components. After melting the polycrystalline samples were annealed in vacuum or in inert argon atmosphere at $T \approx 500–800\,^{\circ}C$ for 2–8 weeks.

The contactless resonator method for examination of the surface electrical resistivity, R_s in a wide temperature range (4.2–300 K) was used [9–11,13]. The electrical surface resistance of the polycrystalline $CeFe_4Al_8$, $ScFe_4Al_8$, $LuFe_4Al_8$, YCr_4Al_8, $CeCr_4Al_8$ and YMn_4Al_8 samples was examined at the frequency of 30 MHz, whereas that of the single crystals of $ScFe_4Al_8$ and YFe_4Al_8 at the frequency of 45 MHz. At the same time, relatively simple geometry of the single crystals of $ScFe_4Al_8$ and YFe_4Al_8 (cylindrical form) makes possible the examination of the electrical resistivity ρ of these samples by the dc four-point technique. Magnetoresistive examination was carried out in magnetic fields up to 0.7 T.

3. Experimental results

The examination of the $ScFe_4Al_8$ and YFe_4Al_8 single crystals has shown that $R_s \sim \rho^{1/2}$ in the whole investigated temperature range which is typical for the normal skin-effect. Taking this observation into consideration, all the $R_s(T)$ dependences for those compounds, for which the geometry does not allow to measure $\rho(T)$, were transformed to the $R_s^2(T)$ form. This fact, in turn, provides the possibility of determining the temperature dependence of ρ in the temperature range at which the anomaly exists. This way of presentation of the experimental results proved to be compatible with the model description of the interaction mechanism which are discussed in this paper.

In this paper, as the reference, characteristic of the NM, the relative quantities are applied: $[R_s^2(H) - R_s^2(0)]/R_s^2(0)$ and $\Delta\rho/\rho(0) = [\rho(H) - \rho(0)]/\rho(0)$, where $R_s(0)$ and $\rho(0)$ are the electrical surface resistance and the resistivity measured in the zero magnetic field, whereas $R_s(H)$ and $\rho(H)$ are the same quantities but measured in magnetic field, respectively.

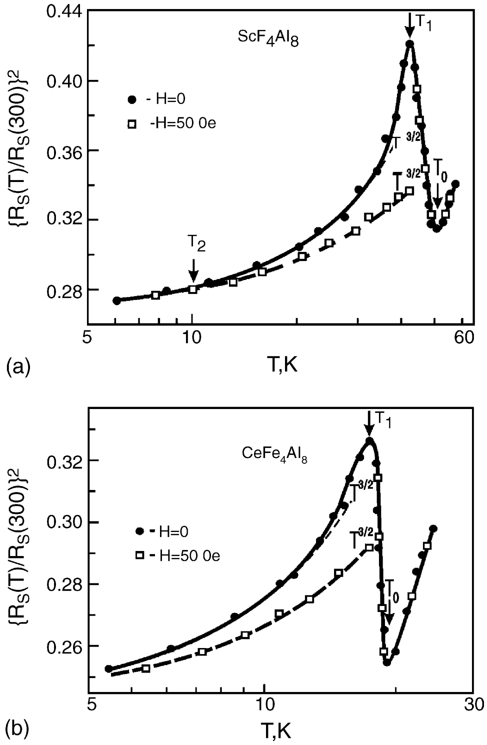

Fig. 2. Temperature dependences of $\{R_s(T)/R_s(300\,K)\}^2$ for the $ScFe_4Al_8$ (a) and the $CeFe_4Al_8$ (b) polycrystalline samples measured with the frequency 10 MHz without the magnetic field and in a magnetic field $H = 50$ Oe.

The temperature dependences of the reduced $R_s^2(T) \sim \rho(T)$ curves as it was defined above for the investigated materials are presented in Figs. 2–5. The field dependence of the magnetoresistivity for $LuFe_4Al_8$ is demonstrated in Fig. 6 (at $T = 40$ K and in applied magnetic fields 0–0.7 T). In turn, in Figs. 7 and 8 one can see the temperature dependences of $R_s^2(T)$ and $\rho(T)$ obtained for the $ScFe_4Al_8$ and YFe_4Al_8 single crystals, respectively, in the different applied current direction. In all these compounds one can see the regions of the anomalous temperature dependences of the electrical resistivity. The observed anomalies are seen in the temperature dependences of the electrical resistive characteristics, which increase from minimum at temperature T_0, reach the maximum at T_1 and then go down to T_2 after which are followed by the regular $\rho(T)$ or $R_s(T)$ dependence. The low magnetic field of 50 Oe suppresses already this anomaly. The characteristic temperatures (T_0, T_1 and T_2) mentioned above, and the values of $[R_s^2(H) - R_s^2(0)]/R_s^2(0)$ and $\Delta\rho/\rho(0)$, taken in a magnetic field $H = 50$ Oe, are listed in Table 1.

3.1. Polycrystalline samples

The $R_s^2(T)/R_s^2(300\ K)$ dependences for the polycrystalline $ScFe_4Al_8$, $CeFe_4Al_8$, $YbFe_4Al_8$, YCr_4Al_8, $CeCr_4Al_8$ and YMn_4Al_8, and $LuFe_4Al_8$ samples are presented in Figs. 2–5 in the temperature range of observed anomalies. The solid points are the data taken at a zero magnetic field, and the squares are those at applied magnetic field $H = 50$ Oe. The solid and dashed lines are the guides for the eye. It follows from these dependences that all these curves are qualitatively similar for all the samples and the largest NM takes place at the temperature T_1. The $CeCr_4Al_8$ compound is a singular exception for which the two maxima at T_1 and T_1' are seen in the $R_s^2(T)/R_s^2(300\ K)$ dependence (Fig. 4b). It is possible that the appearance of these two anomalies is related to the presence of two phases with the same crystal structure but with different stoichiometry. In Fig. 6 the magnetic field dependence of magnetoresistivity for $LuFe_4Al_8$ at $T = 40$ K and in magnetic field up to 0.7 T (in log scale) is shown. This function exhibits the step-like form, with three values of the threshold magnetic field.

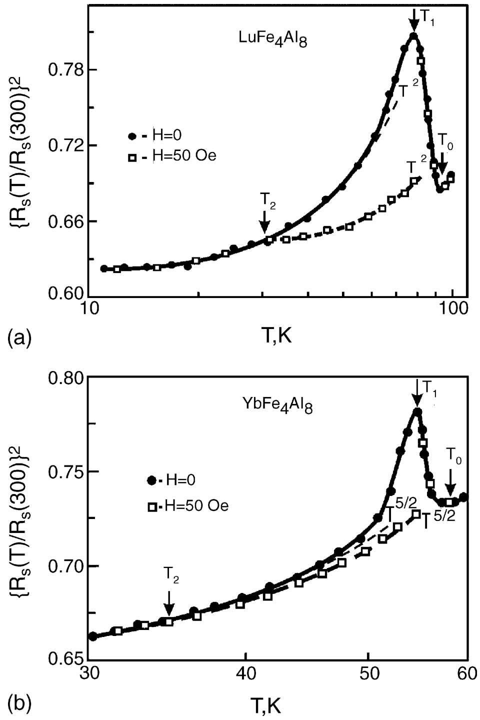

Fig. 3. Temperature dependences of $\{R_s(T)/R_s(300\,\mathrm{K})\}^2$ for the LuFe$_4$Al$_8$ (a) and the YbFe$_4$Al$_8$ (b) polycrystalline samples measured with the frequency 10 MHz without the magnetic field and in a magnetic field $H = 50$ Oe.

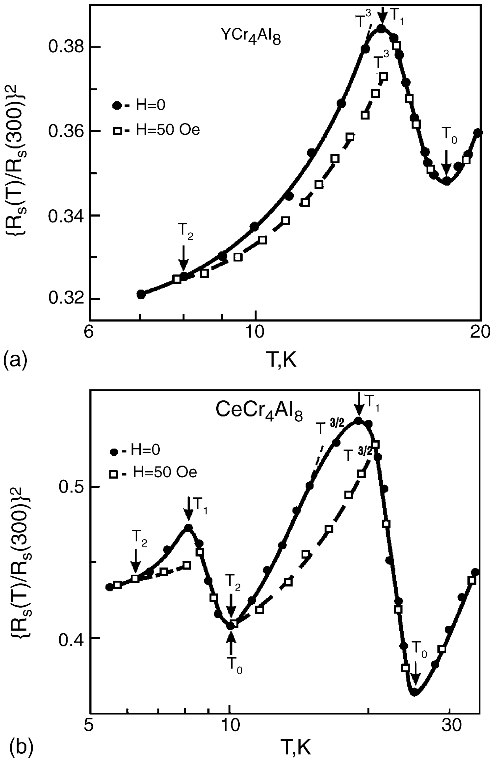

Fig. 4. Temperature dependences of $\{R_s(T)/R_s(300\,\mathrm{K})\}^2$ for the YCr$_4$Al$_8$ (a) and the CeCr$_4$Al$_8$ (b) polycrystalline samples measured with the frequency 10 MHz without the magnetic field and in a magnetic field $H = 50$ Oe.

3.2. Single crystal samples

Typical dependences of the surface resistance $R_s^2(T)$ and the resistivity $\rho(T)$ of the ScFe$_4$Al$_8$ and YFe$_4$Al$_8$ single crystals, in the temperature range of the existence of the magnetoresistive anomalies are shown in Figs. 7 and 8, respectively, for the field directions perpendicular (*a*, *c*) and parallel (*b*, *d*) to the directions of the crystals growth, respectively. It is clear that for the single crystalline samples the $R_s^2(T)$ plots are similar to those for the polycrystalline ones. The influence of the applied magnetic field is the same for both forms of samples. For the ScFe$_4$Al$_8$ single crystal the temperature $T_1 = 43$ K is very close to that for polycrystalline sample, however, the magnitude of the NM is considerably smaller. From the $R_s^2(T)$ measured parallel and perpendicular to the direction of the crystal longer axis, it follows that for these both single crystals the NM is isotropic. It is manifested in equal diminution of the magnitude of the electrical resistivity at $H = 50$ Oe, and also in the existence of the NM in the same temperature range along those directions. The value of $[R_s^2(H) - R_s^2(0)]/R_s^2(0)$ for the ScFe$_4$Al$_8$ single crystal at $T = T_1$ and $H = 50$ Oe amounts to -12%. It is 2.2 times more than the magnitude of $\Delta\rho/\rho(0) \approx -5.5\%$ measured for the same single crystal, but 1.7 times less than this value for the ScFe$_4$Al$_8$ polycrystalline sample. Moreover, the existence range of the NM found in the single crystal measurements is narrower than in the polycrystalline sample (see Table 1). In contrast to the ScFe$_4$Al$_8$, the relations $\Delta\rho/\rho(0)$ and $[R_s^2(H) - R_s^2(0)]/R_s^2(0)$ for the YFe$_4$Al$_8$ show the same magnitude. The comparison of the NM data for the ScFe$_4$Al$_8$ and the YFe$_4$Al$_8$ single crystals shows that the values of $[R_s^2(H) - R_s^2(0)]/R_s^2(0)$ and $\Delta\rho/\rho(0)$ in

Table 1
Typical temperature and magnetoresistance characteristics $[R_s^2(H) - R_s^2(0)]/R_s^2(0)$ and $\Delta\rho/\rho(0)$ in the temperature range where the anomaly and negative magnetoresistivity of the investigated RM$_4$Al$_8$ compounds have been found

Compounds	T_0 (K)	T_1 (K)	T_2 (K)	$[R_s^2(H) - R_s^2(0)]/R_s^2(0)$ (at T_1 in the magnetic field $H = 50$ Oe) (%)	$\Delta\rho/\rho(0)$ (at T_1 in the magnetic field $H = 50$ Oe) (%)
Polycrystalline samples					
CeFe$_4$Al$_8$	20	17	<4.2	−11.5	–
ScFe$_4$Al$_8$	50	43	10	−20.1	–
YbFe$_4$Al$_8$	57	55	35	−7.3	–
LuFe$_4$Al$_8$	97	74	30	−14.3	–
YCr$_4$Al$_8$	18	15	8	−3	–
CeCr$_4$Al$_8$	25	20	10	−6.5	–
YMn$_4$Al$_8$	13	12	8	−3	–
Single crystals					
ScFe$_4$Al$_8$	50	43	25	−12 (⊥ to the growth direction) −12 (‖ to the growth direction)	−5.5 (⊥ to the growth direction) −5.5 (‖ to the growth direction)
YFe$_4$Al$_8$	55	50	30	−1.8 (⊥ to the growth direction) −1.8 (‖ to the growth direction)	−1.8 (⊥ to the growth direction) −1.8 (‖ to the growth direction)

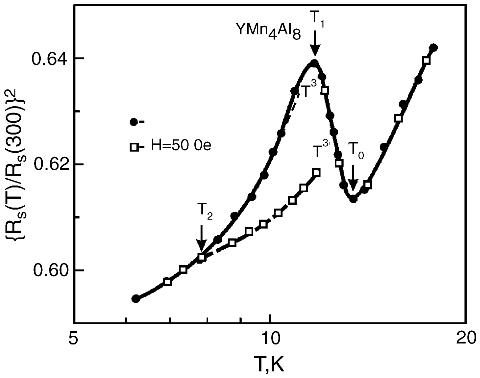

Fig. 5. Temperature dependence of $\{R_s(T)/R_s(300\,K)\}^2$ for the YMn_4Al_8 polycrystalline sample measured with the frequency 10 MHz without the magnetic field and in a magnetic field $H = 50$ Oe.

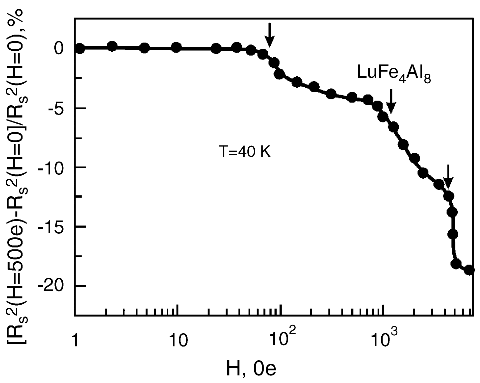

Fig. 6. Magnetic field dependence of $[R_s^2(H = 50\ \mathrm{Oe}) - R_s^2(H = 0)]/R_s^2(H = 0)$ for the $LuFe_4Al_8$ polycrystalline sample measured with the frequency 10 MHz at $T = 40$ K.

the Sc-based compound are 6.7 and 3 times larger, respectively, than those for the Y-based compound.

4. Discussion

The analysis of the experimental results allows to detect the series of the peculiarities in the resistive characteristics of the investigated RM_4Al_8 compounds in the temperature range of the existence of the NM.

The first conclusion is that the NM is observed only for the ternaries in which the M elements are the 3d transition metals (Fe, Cr, Mn) and is absent in the compounds with the noble metals, e.g. Ag and Cu [10].

The indispensable condition for the existence of the NM in the RM_4Al_8 compounds seems to be the presence of the anomaly (maximum) in the temperature dependence of the electrical resistivity at zero magnetic field.

In the Fe-based compounds one observes the coexistence of the NM and antiferromagnetic ordering which appears at higher temperatures ($T_N > 100$ K). In the ternaries with M = Mn and Cr, according to Ref. [14], any magnetic ordering is absent within the range of existence of the NM. Therefore, the antiferromagnetic ordering does not appear the sole reason for the NM.

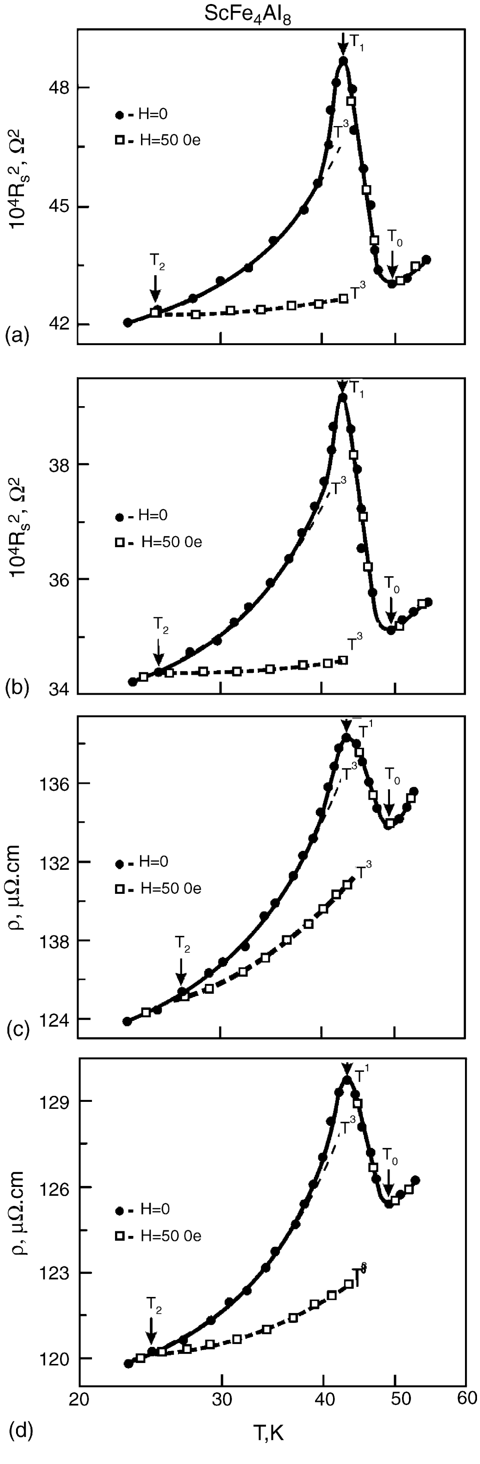

Fig. 7. Temperature dependences of the surface electrical resistance R_s^2 measured with the frequency 45 MHz (a, b) and the electrical resistivity ρ (c, d) of the $ScFe_4Al_8$ single crystal examined perpendicular to (a, c) and parallel (b, d) along the crystal growth direction without the magnetic field and in a magnetic field $H = 50$ Oe.

As mentioned above, the magnetic field dependence of the electrical resistivity for the $LuFe_4Al_8$ polycrystalline sample, taken at $T = 40$ K and in the magnetic field range 0–0.7 T exhibits a complex (step-like) character (Fig. 6).

Let us now consider a question of an influence of a mutual substitution of the R and M atoms on the negative magnetore-

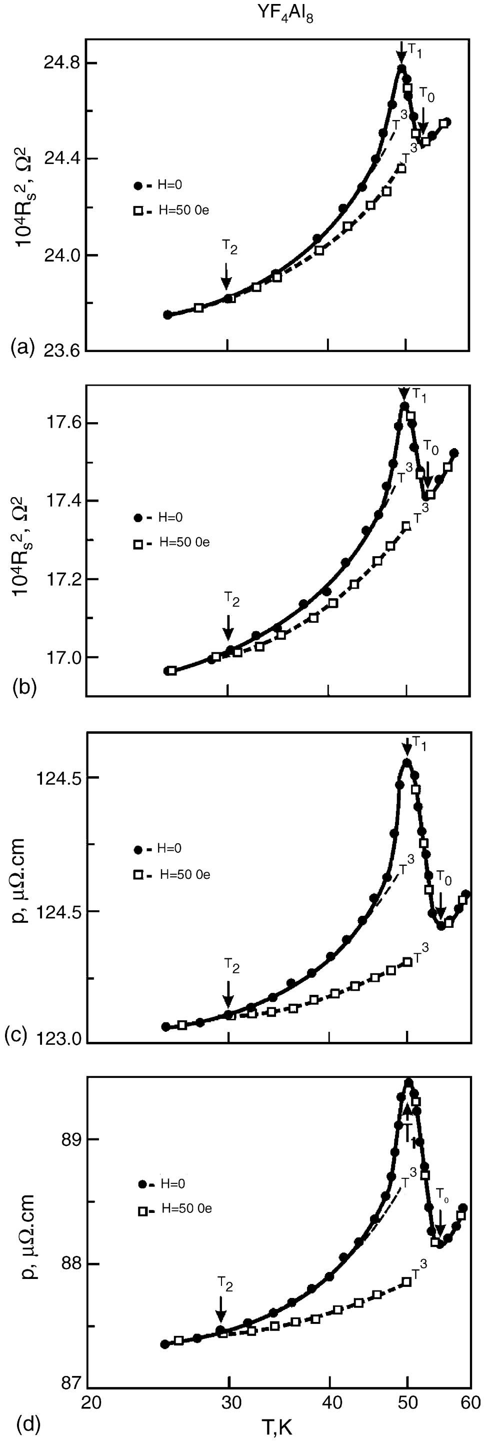

Fig. 8. Temperature dependences of the surface electrical resistance R_s^2 measured with the frequency 45 MHz (a, b) and the electrical resistivity ρ (c, d) of the YFe_4Al_8 single crystal examined perpendicular to (a, c) and parallel (b, d) along the crystal growth direction without the magnetic field and in a magnetic field $H = 50$ Oe.

sistivity in the RM_4Al_8 ternaries. At the beginning we inspect the influence of the various R elements on the examined characteristics.

The careful analysis of the obtained experimental data has shown that the substitution of one rare earth element for another one is a reason for the change in the magnitude of the NM. The dependence of the relative magnetoresistivity at the temperature T_1 in the magnetic field $H = 50$ Oe versus atomic radius of the rare earth atoms is presented in Fig. 9. The upper curve shows this relation for the polycrystals, whereas the lower curve is drawn for the single crystals. It is seen from this figure that as the atomic radius r increases, the relative magnetoresistivity decreases.

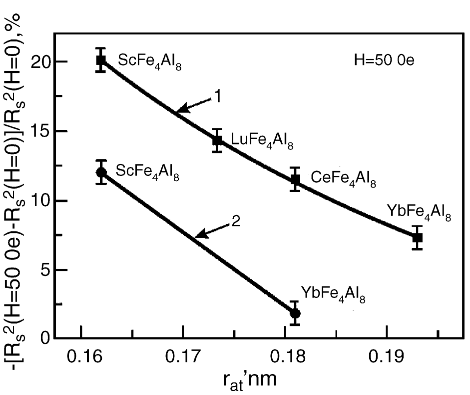

Fig. 9. Magnetoresistance $[R_s^2(H = 50\ \text{Oe}) - R_s^2(H = 0)]/R_s^2(H = 0)$ for the RM_4Al_8 compounds with R = Sc, Lu, Y, Ce and Yb, and M = Fe vs. atomic radius of the rare earth element, R. The upper curve 1 shows this relation for the polycrystals whereas the lower curve 2 is drawn for single crystals.

The influence of the substitution of one transition metal for another, on the magnitude of the NM, observed with the same control parameters (the temperature T_1 and the magnetic field $H = 50$ Oe) versus atomic radius of the transition element is minute if any. Unfortunately, we have only got the data for the two Y compounds containing the different transition metals: YCr_4Al_8 and YMn_4Al_8. For this reason it is difficult to show any regularity in this case. However, one can claim that the NM depends on the atomic radius much weaker when the transition element is substituted instead of the rare earth.

The analysis of the possible mechanisms responsible for the appearance of the negative magnetoresistivity in the RM_4Al_8 compounds allows to say that the basic mechanisms in such systems are the Kondo effect and the spin-glass state formation. This reasoning is based on the following assumptions.

Kondo effect is usually seen in the metallic systems with magnetic impurities [15–17]. With the decrease of temperature, the electrical resistivity in such alloys decreases according to the dependence typical for the nonmagnetic metals, and thus at some characteristic temperature reaches a minimum and then a logarithmic increase appears and finally exhibits the saturation as $T \to 0$. For optimal concentration of the impurities an inter-impurity interaction accompanied by the change of its sign can set up “a local magnetic order” and consequently a state of the spin-glass (SG) in which the direction of the impurity spins is fixed and the electron scattering channel with the spin flip is suppressed [16,17]. The suppression of the dynamic compensation effect of the impurity magnetic moment (Kondo effect) is accompanied by an appearance of the maxima in the temperature dependence of the electrical resistivity T_m (in the present paper—T_1) and in the magnetic susceptibility, T_f [16,17]. After switching on the magnetic field the damping of the maximum in the electrical resistivity is observed. As the result in the temperature range of the existence of this effect appears the negative magnetoresistivity [15–17]. Then T_f becomes the temperature of the transition to the SG state, T_G. Two scenarios can appear: $T_m > T_f$ and $T_m < T_f$. The first inequality is correct for the systems in which $\Delta_c / T_K^{eff} \gg 1$ (where Δ_c is the energy of inter-impurity interaction, T_K^{eff} is the effective temperature of the Kondo system). As an example of such a system one can consider Cu–Mn alloys. At the same time, in some systems, e.g. Cu–Fe or Cu–Co in which the values of Δ_c and T_K^{eff} are of the same order ($\Delta_c / T_K^{eff} \geq 1$), for some concentration of the impurities the inequality $T_m < T_f$ [17] can be fulfilled.

The anomalies in the $\rho(T)$ plot for the RM_4Al_8 compounds similar to those observed for the alloys with magnetic impurities are the following: minimum in this dependence, the range of the logarithmic increase and the degree-like decrease with the diminution of the temperature. This type of behavior of the electrical resistivity and the appearance of the NM allow us to assume the presence in the RM_4Al_8 compounds the Kondo effect and the SG state. The presence of the SG is confirmed by the measurements of the magnetic susceptibility of the single crystals YFe_4Al_8 and $ScFe_4Al_8$ [18,19], in which in the temperature dependence the distinct maxima have been detected in these temperature ranges in which the anomalies in the $\rho(T)$ plot exist. Strong dependence of the singularities on the cooling conditions of the single crystals inclined the authors to assume that in this temperature range the transition into the region of the cluster SG takes place. The comparison of the temperature of the maxima of the electrical resistivity and the magnetic susceptibility shows that for the YFe_4Al_8 single crystal $T_m > T_f$ ($T_m = 50$ K, $T_f = 38$ K [18,19]). At the same time for the $ScFe_4Al_8$ single crystal $T_m < T_f$ ($T_m = 43$ K, $T_f = 60$ K in Refs. [18,19] and $T_f = 135$ K in Ref. [12]).

The Kondo effect and the SG state in RM_4Al_8 compounds, apparently, are related to the existence in these compounds some number of the transition element atoms located not only in the 8f sites but also in the 8j positions, improper for them. According to the references (see, e.g. [20,21]) such a situation frequently exists in these systems. However, due to density functional theory calculations for YFe_4Al_8 there is no need for a second Fe sublattice with an arbitrary phase difference to explain the neutron diffraction results [22].

Let us inspect separately systems with the Fe and Mn examined by us. According to the Mössbauer effect investigations [12] the Fe atoms distributed on the 8j sites could exist in a paramagnetic state, exhibiting a local magnetic moment and could be effective as the magnetic impurity as is observed in the Kondo systems. Here, one should note that the principal contribution to the electrical conductivity of the RM_4Al_8 compounds is due to the free charge carriers of 3p(Al)-band [23]. The appearance of the Fe atoms at the some positions of the Al atoms is a reason for the scattering of the 3p electrons with an inversion of the spin which is reflected in the maximum in the $\rho(T)$ plot of the investigated samples. Further, it is necessary to point out that the localization of the iron at 8j position is responsible for the appearance of the competing antiferromagnetic 8f–8f and ferromagnetic 8f–8j exchange interactions at low temperature [24]. The result of such a competition is the frustration of the magnetic moments of the transition metal which is a reason for the formation of the spin-glass state. Particularly, for this reason the temperature dependence of the magnetic susceptibility $\chi(T)$ of the YFe_4Al_8 and $ScFe_4Al_8$ in the range of the electrical resistivity anomalies strongly depends on the thermal history of the samples [18,19]. According to above discussion it can be stated that various temperatures of the transitions in the SG state for the $ScFe_4Al_8$, obtained in different papers ($T_f = 60$ K in Refs. [18,19], $T_f = 135$ K in Ref. [12]), and also the existence the $T_m < T_f$ relation for the $ScFe_4Al_8$, which is different from that observed for the YFe_4Al_8 ($T_m > T_f$), can be related to the various degree of the Fe and the Al atoms disorder in these alloys. At the same time we assume that the stoichiometry of the YFe_4Al_8 samples which have been investigated by us and in Refs. [18,19] are close to each other. Such a conclusion is justified by the following facts. According to Ref. [20] a small deviation from 1:4:8 stoichiometry in RFe_4Al_8 compounds in the direction of the higher Fe concentration results in considerable decrease of the magnetic transition temperature. For example, in YFe_4Al_8 $T_N = 180$ K, and in $YFe_{4.2}Al_{7.8}$ $T_N = 100$ K [20]. Therefore, the Néel temperature can be an indication of the perfection of the RFe_4Al_8 samples. (the compounds with higher T_N have smaller deviation from the 1:4:8 stoichiometry in direction of the higher concentration of the Fe). In our case the T_N of the YFe_4Al_8 and $ScFe_4Al_8$ single crystals obtained from resistance measurements amount to 100 and 210 K, respectively. Comparing the data for YFe_4Al_8 with Refs. [18,19] it is seen that for the YFe_4Al_8 our T_N is in agreement with the Néel temperature determined in the magnetic measurements. It can prove that this sample exhibits not too large deviation from stoichiometry 1:4:8 and corresponds to the 1:4.2:7.8 composition according to Ref. [20]. For the $ScFe_4Al_8$ compound the situation is quite the opposite. T_N determined in the present research is higher than in Refs. [12,18,19]. It could be an indication that at present, the investigated sample is closer to the 1:4:8 than the samples investigated in Refs. [12,18,19]. The latter samples exhibit a deviation from 1:4:8 stoichiometry towards a higher concentration of the Fe on the 8j sites, i.e. one can claim that the YFe_4Al_8 samples investigated in different papers have fairly close stoichiometry, resulting in fairly equal temperatures T_f and T_m. At the same time the stoichiometry of the samples of the $ScFe_4Al_8$ in the present paper and in Refs. [12,18,19] differs strongly and therefore the compound shows different T_f and T_m values. According to Refs. [15–17] in the Kondo systems the increase of the magnetic impurity concentration causes an increase of T_f and T_m as well. Thus, for the $ScFe_4Al_8$ compound as for the YFe_4Al_8 compound, one can expect the fulfilling of the relation $T_m > T_f$. However, it is not excluded that in the $ScFe_4Al_8$ alloy the energy of inter-impurity interactions and the effective Kondo temperature of the Kondo system causes $T_m < T_f$ inequality. The final rectification of this problem needs a further examination.

As it is known in the RMn_4Al_8 long range magnetic ordering is absent [25–27]. In this case the magnetic state of the Mn sublattice is determined by the interatomic Mn–Mn separation, by the exchange interactions with the closest neighbors and by the temperature [25]. According to the data of Refs. [25–27] the separation $d_c \approx 2.7$ Å appears to be critical. At $d < d_c$ the Mn atoms do not have the local magnetic moment. In the YMn_4Al_8 compound with the exact stoichiometry the Mn atoms occupy only the 8f sites, forming quasi-unidimensional chain along the *c*-axis with the separation $d_{ff} = 2.55$ Å $< d_c$ [25]. In such a case the localized magnetic moments of the Mn atoms do not exist. Due to the small rearrangement of the Mn and Al atoms, the former atoms appear also in the 8j positions. The separation between the j–j sites of the Mn atoms amounts now to the value $d_{jj} = 2.77$ Å $> d_c$ [28] and the Mn atoms in 8j sites exhibit local magnetic moment. Now they can play a role of magnetic impurities and at the low temperature they can cause the Kondo effect. Recently, it has been shown that in the RMn_4Al_8 compounds, in spite of the absence of the long range magnetic interactions the strongly developed spin fluctuations (SF) are present [25–27]. At low temperatures the fluctuations of the Mn moment become slower resulting supposedly in forming of the dynamical SG [26]. Thus the last phenomenon could be responsible for the damping of the Kondo effect and the decrease of the electrical resistivity with a diminution of the temperature observed by us in YMn_4Al_8.

In this way, we deal with the situation in which different mechanisms of the internal interactions in the RM_4Al_8 systems with the iron and the manganise lead to the same final result—appearance of the Kondo effect and SG. As far as the systems with the chromium are concerned, at present, unfortunately, we have not sufficient experimental results, for the detailed analysis.

The law governing the decrease of $\rho(T)$, after reaching the maximum in the $\rho(T)$, is dependent on different factors and can be variable. The decrease could be described by the Fermi liquid mechanism according to $\sim T^2$ dependence [15], however the same formula can be applicable because of the spin-glass formation [29]. The $\sim T^3$ law appears as the result of the s-d scattering due to phonons [30]. According to Ref. [31] for the spin-glass systems, the $T^{3/2}$ law can also be detected. It is possible that simultaneous action of these mechanisms causes withdrawn the degree of the electrical resistivity to decrease as a T^n where n varies between 1.5 and 3.0. For all the samples measured in the present work the resistivity increases in the logarithmic way starting from the temperature at which the resistive characteristics commence to increase, which is specific for each material. Then this increase changes as the T^n dependence when the temperature is lowered and the power n aims at the range 1.5–3.0 for all materials investigated here.

Taking into account what was presented above one can understand why after some time (a few months) or as the result of the multiple thermo-cycling the magnitude of the NM decreases, and even it completely vanishes. It could be connected with the fact that such a process involves the equilibrium in the primarily disordered systems in which at the beginning the transition metal atoms occupy the improper positions. For the examination of this conclusion we prepared perfect samples of the investigated compounds, which were annealed at 500 °C during more than 8 weeks. For all these samples the smooth temperature dependence of the electrical resistivity, without any anomaly, is observed.

These facts presented above allow to propose three basic scenarios for the explanation of the observed behavior. The first one can be considered when the stoichiometric but nonbalanced sample is discussed. Then the phenomenon of the NM is observed, which in time or after thermal treating decreases down to its full decay. The second one is valid when we are dealing with the nonstoichiometric sample with a small excess of the transition metal. In this case not only the 8f sites are fully occupied, but also a part of the 8j positions is occupied by the transition metal atoms. Such samples exhibit the NM in the stable and the certain way. The third scenario concerns the situation when the originally stoichiometric and being in the equilibrium alloys do not exhibit the NM, i.e. the atoms of the transition element, which can be treated as the magnetic impurities are absent at the 8j positions.

It has been shown above that for the YFe_4Al_8 and $ScFe_4Al_8$ single crystals the $R_s \sim \rho^{1/2}$ dependence is followed in the whole investigated temperature range. From that, in turn, it results that it is reasonable to assume the equality of the relative magnitude of the NM if the measurements are performed with the dc and ac currents. In reality, $[R_s^2(H) - R_s^2(0)]/R_s^2(0)$ and $\Delta\rho/\rho(0)$ plots are the same for the YFe_4Al_8 single crystal, whereas for the $ScFe_4Al_8$ single crystal 2.2 times diminution of the magnetoresistivity in the magnetic field is detected and it is more pronounced when the ac current is applied. Such behavior can be easily explained if one assumes that the surface layer which we are dealing with performing the measurements with the ac current in the $ScFe_4Al_8$ single crystal, is more affected by the admixture of the Fe atoms to the 8j positions than the bulk material. At the same time, we have the reasons to claim that the YFe_4Al_8 single crystal has more homogeneous distribution of the Fe atoms at the 8j sites. Arguing, according to the same lines, one can suggest an explanation of the higher NM in the polycrystalline sample in relation to the single crystals, due to the more expanded surface. Taking into consideration the arguments provided above, the step-like dependence of $[R_s^2(H) - R_s^2(0)]/R_s^2(0)$ for the $LuFe_4Al_8$ polycrystalline sample can be related to the nonhomogeneous distribution of the Fe atoms at the 8j sites in the alloy. Therefore, for suppressing the electron scattering with a spin reversal process in the various fractions of the bulk material the various energy of the magnetic field is necessary.

Recent investigations show that step-like $R_s(H)$ structures can be seen within the T_1–T_2 temperature interval and around T_1 only. Within the T_0–T_1 temperature interval $R_s(H)$ structures have smooth character. This may indicate that some kind of phase separation processes are developed at temperatures below T_1.

5. Conclusion

The analysis of the experimental data and the discussion of the possible mechanisms of the interactions of the individual sublattices and the charge carriers in the RM_4Al_8 compounds (where R—nonmagnetic rare earth as well as Y, Sc and M—transition metal) allows us to suppose that the main reason for the appearance of the negative magnetoresistivity is the Kondolike interaction. The important factor is also the spin-glass state frequently observed in these ternaries. The considered phenomena can result from the crystallographic imperfection (nonstoichiometry and interchange of the site occupation) of these alloys. In the stoichiometric samples with the ideal distribution of the atoms in the lattice, neither the NM nor the SG state appears. The corresponding measurements can be used as the indication of the sample quality.

The experiments show that when the NM is observed, its overall magnitude increases in case when the atomic radius of the rare earth atom in the RM_4Al_8 compounds is smaller. The NM practically does not depend upon the atomic radius of the transition element. It shows the dominant significance of the rare earth atoms in the interactions responsible for the scattering mechanism of the charge carriers in the RM_4Al_8 systems.

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