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Electrical resistivity of $Lu(M_{1-x}Al_x)_2$ (M≡Co, Ni)

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Abstract

The behaviour of the electrical resistivity ρ of $Lu(Co_{1-x}Al_x)_2$ and $Lu(Ni_{1-x}Al_x)_2$ has been studied as a function of the aluminium concentration, temperature and applied magnetic field. In the Pauli paramagnet $LuCo_2$ a larger value of the residual resistivity ρ_{res} than in $LuNi_2$ and the tendency to saturation of the $\rho(T)$ dependence at temperatures $T > 150$ K are observed. In $LuNi_2$, in which the exchange enhancement has not been observed, residual resistivity increases monotonically with increasing aluminium substitution on nickel sites. The concentration dependence of ρ_{res} in $Lu(Co_{1-x}Al_x)_2$ shows a maximum at $x = 0.08$ near the critical concentration for the onset of ferromagnetism. The difference in behaviour of these two systems is explained by a large contribution to the electrical resistivity from spin fluctuations and by a change in this contribution with changes in the aluminium content, temperature and magnetic field.

1. Introduction

The Laves phase compound $LuCo_2$, as well as other RCo_2 (R≡Y, Sc) compounds in which the R ion has no intrinsic magnetic moment, are exchange-enhanced Pauli paramagnets [1, 2]. The temperature dependence of the electrical resistivity of YCo_2 shows a tendency to saturation at high temperatures ($T > 150$ K) [3, 4]. The temperature dependences of the magnetic susceptibility of these three compounds have a broad maximum in the temperature region 200–400 K [2]. The peculiarities of transport and magnetic properties of RCo_2 (R≡Y, Lu, Sc) originate from the electronic structure of these compounds. The Fermi level of YCo_2 lies on a sharply falling part of the density of states (DOS) curve, as a function of energy [5]. For $LuCo_2$ a detailed calculation of the electronic structure has not been performed. However, a similar $N(E)$ dependence near the Fermi level can be expected in $LuCo_2$ as in YCo_2, because these compounds have similar magnetic properties.

A partial substitution of aluminium for cobalt in $LuCo_2$ leads at first ($x < 0.08$) to an exhibition of itinerant metamagnetism, and then to the appearance of ferromagnetism with a magnetic moment per Co atom $\mu_{Co} \approx 0.63\mu_B$ [6–9].

In this paper, the experimental results of electrical resistivity measurements in the temperature range 4.2–300 K and under applied magnetic fields

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up to 7.5 T are reported for $Lu(Co_{1-x}Al_x)_2$ and $Lu(Ni_{1-x}Al_x)_2$. The $LuNi_2$ compound is a Pauli paramagnet, in which (as in YNi_2 [5]) the Fermi level probably lies on a part of the $N(E)$ curve with a very low local DOS. The difference in the electronic structure and magnetic properties of these two systems should lead to the difference in their transport properties.

2. Experimental details

The $Lu(Co_{1-x}Al_x)_2$ and $Lu(Ni_{1-x}Al_x)_2$ samples with x up to 0.10 were prepared by melting lutetium (99.9% pure) and cobalt, nickel and aluminium (99.99% pure) in an arc furnace. The ingots were annealed in evacuated quartz tubes at 850 °C for 1 week. X-ray diffraction and metallographic analysis confirmed that $Lu(Co_{1-x}Al_x)_2$ with x up to 0.09 and $Lu(Ni_{1-x}Al_x)_2$ with x up to 0.10 were single phases. At an aluminium content $x > 0.09$ in the $Lu(Co_{1-x}Al_x)_2$ compounds foreign phases were observed. The electrical resistivity was measured with the four-probe method on prismatic specimens of about $1 \times 1 \times 6$ mm^3. The error in determination of the specific resistivity was about 5% and its relative change with temperature or in a magnetic field was not larger than 0.1%. The temperature was measured using a germanium resistance thermometer, as well as a copper–constantan thermocouple.

3. Results and discussion

3.1. $LuCo_2$ and $LuNi_2$

It can be seen from Fig. 1 that the temperature dependence of the electrical resistivity of the $LuCo_2$ compound shows the tendency to saturation at temperatures $T > 150$ K as for YCo_2 [4]. In the case of the YCo_2 compound the non-linear $\rho(T)$ dependence at $T > 150$ K was explained by two causes: (a) the scattering of electrons on the spin fluctuations [3]; (b) the influence of the fine structure of the $N(E)$ curve near the Fermi level [4].

Figure 1 also presents the $\rho(T)$ dependence for the isostructural $LuNi_2$ compound. This dependence is typical for the metallic systems, in which the dominant temperature-dependent contribution in the electrical resistivity is a contribution from the electron–phonon interaction. The broken curve in Fig. 1 shows the part of the resistivity of the $LuCo_2$ compound after subtraction of the phonon contribution $\rho(T) - \rho_{ph}(T)$. $\rho_{ph}(T)$ is obtained from the $\rho(T)$ dependence measured on the $LuNi_2$ sample. An increase of the temperature causes a decrease of the $\rho(T) - \rho_{ph}(T)$ part after a maximum at $T = 150$–200 K. In the model of spin fluctuations the similar temperature dependence of the resistivity is determined by the interaction between the spin fluctuation modes at high temperatures [10]. It draws attention to the fact that the value of ρ_{res} for $LuCo_2$ is larger than that for $LuNi_2$. As shown in ref. 11, the residual resistivity for the exchange-enhanced Pauli paramagnet YCo_2 also exceeds the ρ_{res} value for the isostructural YAl_2 compound, in

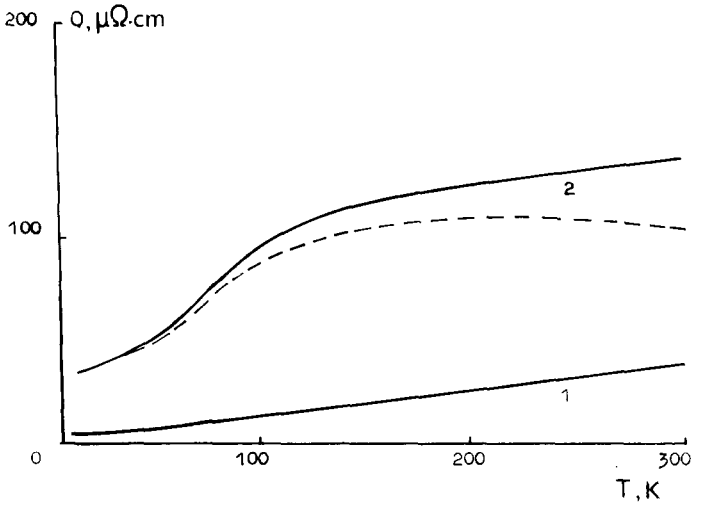

Fig. 1. Temperature dependences of the electrical resistivity ρ for $LuNi_2$ (curve 1) and $LuCo_2$ (curve 2): – – –, $\rho(T) - \rho_{ph}(T)$ for $LuCo_2$.

which the exchange enhancement could not exist. This difference can also be determined by the difference in electronic structure of the YCo_2 and YAl_2 compounds.

The location of the Fermi level of RCo_2 (R ≡ Y, Lu, Sc) on the sharply falling part of the $N(E)$ curve can lead to a situation when the presence of crystal structure defects and inhomogeneity of the chemical composition over the volume of the sample cause the fluctuations of the local DOS and the spin fluctuations. The latter can determine the additional magnetic contribution to the scattering of the conduction electrons and the larger ρ_{res} value in comparison with compounds in which the Stoner exchange enhancement factor is small. The similar difference between the ρ_{res} values observed in $LuCo_2$ and $LuNi_2$ can take place in other systems.

3.2. Lu(Co,Al)$_2$ and Lu(Ni,Al)$_2$

The $\rho(T)$ dependences for the $Lu(Ni_{1-x}Al_x)_2$ are given in Fig. 2. The substitution of aluminium up to $x = 0.10$ causes the monotonic increase of ρ_{res} while the phonon contribution $\rho_{ph}(T)$ practically does not change with increasing aluminium concentration. This behaviour is due to the increase of the concentration of scattering centres. Another picture is observed in the $Lu(Co_{1-x}Al_x)_2$ compounds. As can be seen in Fig. 3, the partial substitution of aluminium for cobalt causes at first an increase in the residual resistivity and then ($x > 0.08$) its decrease. Moreover, at $x > 0.08$ the $\rho(T)$ dependences show an anomaly accompanying the appearance of magnetic order. The magnetic ordering temperatures T_c, which are determined from $\rho(T)$ measurements, have a good agreement with data from the magnetic measurements [6]. The values of the critical concentration for the onset of ferromagnetism ($x_c \approx 0.085$), determined from these two methods, are also equal.

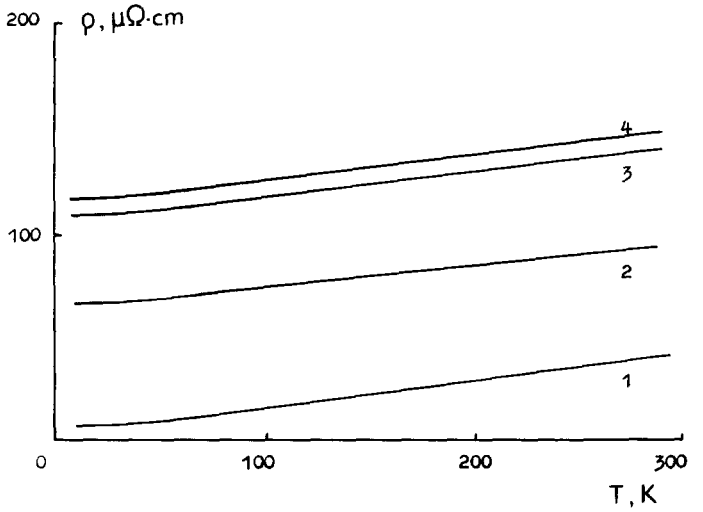

Fig. 2. The $\rho(T)$ dependences for $Lu(Ni_{1-x}Al_x)_2$: curve 1, $x=0.0$; curve 2, $x=0.03$; curve 3, $x=0.06$; curve 4, $x=0.10$.

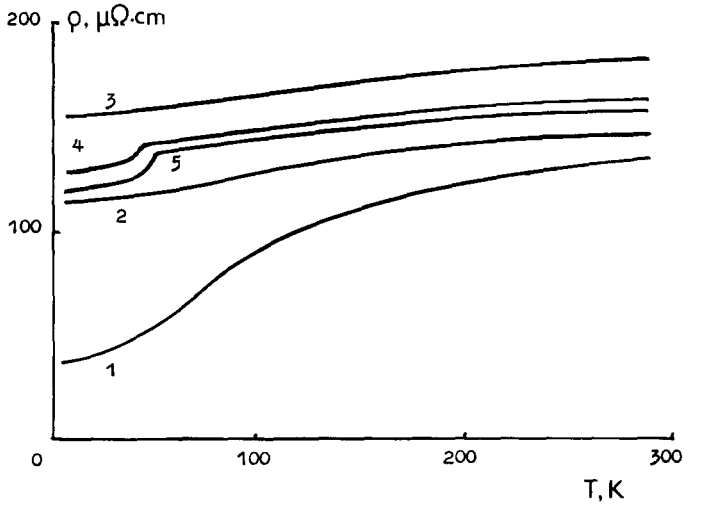

Fig. 3. The $\rho(T)$ dependences for $Lu(Co_{1-x}Al_x)_2$: curve 1, $x=0.0$; curve 2, $x=0.02$; curve 3, $x=0.08$; curve 4, $x=0.085$; curve 5, $x=0.10$.

The concentration dependences of ρ_{res} and ρ at 300 K (denoted by $\rho(300)$) for $Lu(Ni_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Al_x)_2$ compounds are given in Fig. 4. The shaded area in Fig. 4 represents the additional contribution in the residual resistivity, which is probably determined by scattering of the electrons on the spin fluctuations. This assumption is confirmed by results obtained from NMR measurements [12]. As is shown in ref. 12, in $Lu(Co_{0.92}Al_{0.08})_2$ a coexistence of non-magnetic and magnetic ($\mu_{Co} \approx 0.63\mu_B$) states of the cobalt atoms is observed. This means the presence of spin density fluctuations

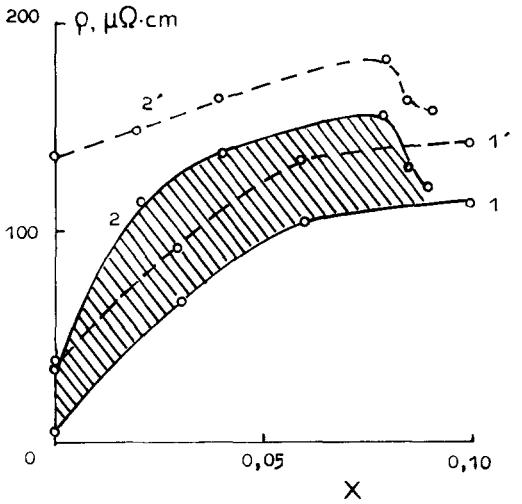

Fig. 4. Concentration dependences of the ρ_{res} (curve 1) and $\rho(300)$ (curve 1') values for $Lu(Ni_{1-x}Al_x)_2$ and of ρ_{res} (curve 2) and $\rho(300)$ (curve 2') for $Lu(Co_{1-x}Al_x)_2$.

in the volume of the sample. As follows from Fig. 4, just at $x=0.08$ the maximum ρ_{res} is observed. The decrease in residual resistivity at $x>0.08$ is probably determined by the suppressing of spin fluctuations on magnetic ordering and also by the change of electronic structure with the aluminium substitution. As noted above, the electronic structures of $LuCo_2$ and YCo_2 can be expected to be similar, because these compounds exhibit similar magnetic properties. As follows from the calculation of the electronic structure for YCo_2 [5] and for the ordered compound $Y(Co_{0.75}Al_{0.25})_2$ [13] the behaviour of these compounds cannot be treated with a rigid band model [14]. The addition of aluminium to YCo_2 broadens a narrow peak in the DOS near the Fermi level of YCo_2 and changes the DOS value at the Fermi level [13].

The concentration dependence of the residual resistivity obtained in the present work for $Lu(Co_{1-x}Al_x)_2$ compounds correlates with the behaviour of the coefficient γ of the linear term of the low-temperature specific heat. As is shown in ref. 15, the increase in the aluminium content leads to a non-monotonic change of the γ value with a maximum at $x=0.08$. A sharp decrease in the γ value is observed at $x>0.08$, where the ferromagnetic state is stabilized. The concentration dependence of the γ value is explained by spin fluctuations and by changes in the DOS at the Fermi level.

At $x<0.08$ in the $Lu(Co_{1-x}Al_x)_2$ compounds the metamagnetic transitions have been observed by measuring the magnetization and forced magnetostriction [16]. It can be expected that the application of an external magnetic field as well as the spontaneous magnetic ordering is accompanied by the suppression of spin fluctuations and, consequently, by a decrease in the electrical resistivity. This behaviour is really observed with the longitudinal magnetoresistivity measurements. As can be seen from Fig. 5, which shows the field dependence of $\Delta\rho/\rho$ for $x=0.08$ at $T=4.2$ K, a sharp decrease of

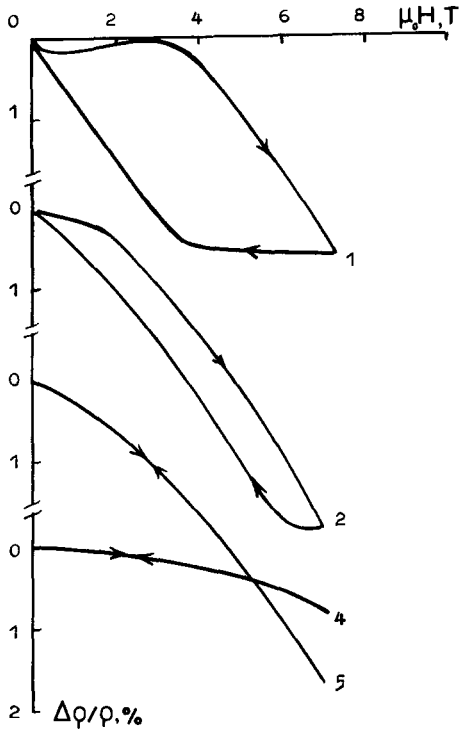

Fig. 5. Longitudinal magnetoresistivity $\Delta\rho/\rho$ *vs.* the field for $Lu(Co_{0.92}Al_{0.08})_2$ at various temperatures T: curve 1, 4.2 K; curve 2, 20 K; curve 3, 40 K; curve 4, 80 K.

the electrical resistivity is observed as the field is increased to the critical value. The increase of the temperature leads to the decreasing of the critical field and of the width of the hysteresis.

4. Conclusion

The isostructural $LuCo_2$ and $LuNi_2$ compounds, the first of which is an exchange-enhanced Pauli paramagnet and in the second of which the Fermi level crosses the $N(E)$ curve at very low DOS, have significantly different temperature dependences of their electrical resistivity. For $LuCo_2$ a large contribution to the electrical resistivity from scattering on the spin fluctuations is observed. This contribution causes the greater value of residual resistivity in $LuCo_2$ than in $LuNi_2$ and a tendency to saturation at temperatures $T > 150$ K. It can be expected that a similar difference in the transport properties should be observed in other metallic systems with a similar difference in their electronic structures. The partial substitution of aluminium for nickel in $LuNi_2$ leads to monotonic increasing of the ρ_{res} value due to the increasing of the concentration of scattering centres. However, the concentration dependence of the ρ_{res} value for the $Lu(Co_{1-x}Al_x)_2$ system shows a maximum

ρ_{res} at $x=0.08$ and its decrease at $x>0.08$, where ferromagnetic order is stabilized. The observed behaviour of the electrical resistivity in these compounds is determined by the change in the contribution from the spin fluctuations and also by the change of electronic structure with increasing aluminium content. Suppression of the spin fluctuations due to the spontaneous magnetic ordering as well as at the metamagnetic transition in the d electron system produced by an applied magnetic field causes the decreasing of the electrical resistivity.

Acknowledgments

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