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Electrical resistivity of RNi_4Al and RNi_4Cu

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Comparative studies of the temperature dependence of resistivity for RNi_4X (X = Al, Cu; R = Y or rare earth) compounds are presented. These results are also compared to the previously obtained for RNi_4B. The ordering temperatures are well identified on the $\rho(T)$ curves. The residual resistivity ρ_0 of RNi_4Al is several times higher than those of RNi_4Cu and RNi_4B. The YNi_4X compounds are included in these studies as the nonmagnetic isostructural reference materials. The phonon contribution has been determined for both Y-based compounds employing the Bloch–Grüneisen formula. The $CeNi_4X$ compounds show a shallow minimum in $\rho(T)$ at low temperatures (about 15 K). This anomaly has been ascribed to the Kondo impurity-like behaviour. For the other rare earths, the $\rho(T)$ dependences below T_C have been analyzed assuming the scattering on magnons as the predominant mechanism.

Key words: *rare earth compound; electrical resistivity*

1. Introduction

The analysis of the temperature dependence of the electrical resistivity in the intermetallic compounds is a powerful tool to get information on the intrinsic properties of these materials. Depending on the temperature range considered, one can conclude on the scattering of electrons on the thermal excitations of the lattice or on magnons.

In this paper, we present the analysis of the $\rho(T)$ curves for RNi_4Cu and RNi_4Al compounds (R = Y or rare earth), both crystallizing in the hexagonal $CaCu_5$-type of structure. For the sake of comparison, our previous studies on the RNi_4B series ($CeCo_4B$-type of structure) [1] are recalled within this paper.

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2. Results and discussion

We assume that the Matthiessen rule is fulfilled, i.e., the temperature dependence of resistivity can be treated as a sum of the involved contributions:

$$\rho(T) = \rho_0 + \rho_{ph}(T) + \rho_m(T) \tag{1}$$

where ρ_0 is the residual resistivity, $\rho_{ph}(T)$ represents the phonon term and $\rho_m(T)$ is the magnetic contribution.

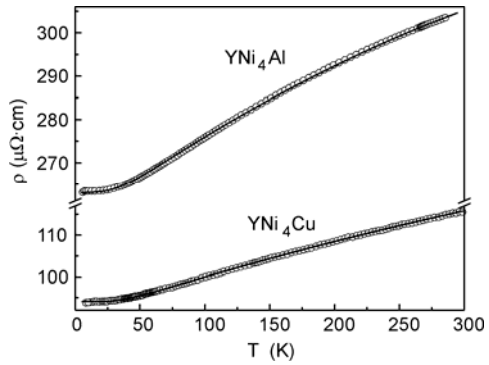

Fig. 1. The $\rho(T)$ curves of the nonmagnetic YNi_4Cu and YNi_4Al compounds. The solid lines represent fits with Eq. (2)

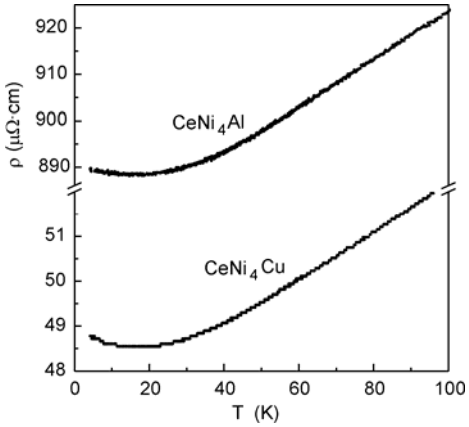

Fig. 2. The $\rho(T)$ dependences for $CeNi_4Cu$ and $CeNi_4Al$. Shallow minima due to the Kondo impurity like behaviour are visible at low temperatures

Figure 1 shows the $\rho(T)$ curves measured for the nonmagnetic YNi_4Cu and YNi_4Al compounds. The solid line is a fit to the formula:

$$\rho(T) = \rho_0 + 4R\Theta_D\left(\frac{T}{\Theta_D}\right)^5 \int_0^{\Theta_D/T} \frac{x^5\,dx}{\left(e^x - 1\right)\left(1 - \mathrm{e}^{-x}\right)} - KT^3 \tag{2}$$

where the second right-hand term is the Bloch–Grüneisen relation and the last term denotes the Mott scattering. This term is mainly reflected in the curvature visible in the range from about 50 K up to room temperature. For YNi_4Al we have obtained the Debye temperature $\Theta_D = 204$ K, the constants $R = 0.165$ μΩ·cm/K and $K = 2.25\times10^{-7}$ μΩ·cm/K^3. In the case of YNi_4Cu, the parameters are: $\Theta_D = 235$ K, $R = 0.08$ μΩ·cm/K and $K = 0.5\times10^{-7}$ μΩ·cm/K^3. In the previous studies on YNi_4B, we have determined $\Theta_D = 240$ K, $R = 0.109$ μΩ·cm/K and $K = 0$ [1]. These values of the Debye temperature suggest that the shortest phonon wavelength occurs for YNi_4B. It seems to correlate well with the structural features, namely the lattice constant a is similar for all the RNi_4X (X = Al, Cu, B) series ($a \approx 5$ Å), whereas in the c direction the R–R separation is in average 4 Å, 3.98 Å and 3.42 Å for X = Al, Cu and B, respectively.

The $\rho(T)$ dependences of the $CeNi_4Cu$ and $CeNi_4Al$ compounds are plotted in Fig. 2. The Cu-based compound exhibits a shallow minimum at low temperatures, which resembles the case of the Al- [2] and B-based [3] samples. It is ascribed to the Kondo impurity-like effect, developed by a very small amount of Ce precipitates with well localized magnetic moment. In general, it is also possible that the $CeNi_4X$ (X = B, Al, Cu) compounds are intermediate between the mixed valence and the Kondo lattice systems.

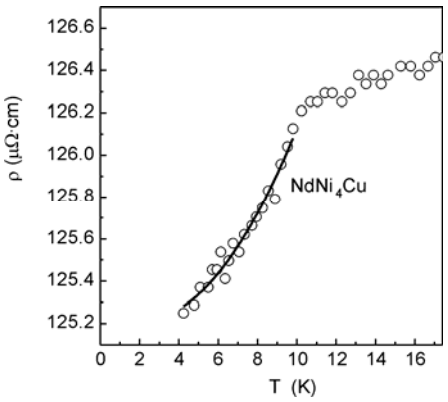

Fig. 3. The magnetic contribution $\rho_m(T)$ for $NdNi_4Cu$ fitted with Eq. (3). T_C is visible at ca. 11 K

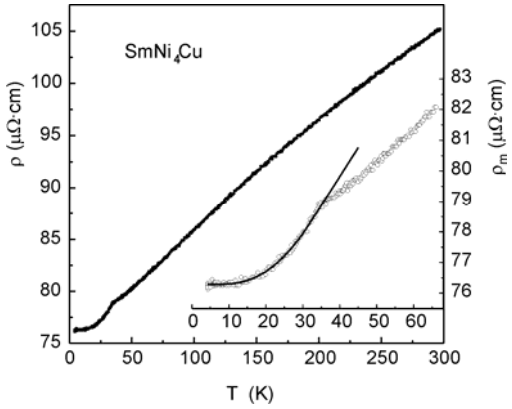

Fig. 4. The $\rho(T)$ curve of $SmNi_4Cu$ in the full temperature range. Inset: the magnetic part $\rho_m(T)$ fitted with Eq. (3) below the phase transition

$NdNi_4Al$ and $NdNi_4Cu$ exhibit transitions to the ferromagnetic order at 6 K and 11 K, respectively. These temperatures are well visible in the resistivity (Fig. 3) and coincide well with the previous magnetometric findings [4]. Below T_C the main contribution to the resistivity comes from the scattering on the spin wave excitations and is usually represented by a $\rho_m \sim T^2$ dependence. However, in the presence of a strong magnetic anisotropy a gap Δ can be developed in the magnon spectrum ($E_k = \Delta + Dk^2$) and $\rho_m(T)$ takes the form [5]:

$$\rho_m(T) = \rho_0 + m\Delta T e^{-\Delta/T}\left(1 + 2\frac{T}{\Delta}\right) \tag{3}$$

where m is a material constant. The fit below T_C (Fig. 3) provides Δ = 7.7 K for $NdNi_4Cu$, it was 19 K for $NdNi_4B$ [1] and the spread of the experimental points does not allow a reasonable fit in the case of $NdNi_4Al$. Figure 4 demonstrates that Eq. (3) operates perfectly in the case of $SmNi_4Cu$ providing Δ = 40 K. Sm-based compounds are usually characterized by a strong magnetic anisotropy. We have observed the coercive field of 2.8 T for $SmNi_4Cu$ and 7 T for $SmNi_4B$ [6]. The latter compound has shown the energy gap Δ = 42 K. Hence, it is evident that there is a correlation between the parameter Δ and the coercive field. The $NdNi_4X$ compounds have shown

H_C of the order of 10^{-3} T. In the case of $GdNi_4X$ the magnetic anisotropy is negligible because Gd is in the S state (orbital number $L = 0$). It is corroborated by the resistivity measurements, which reveal that the temperature variation below T_C fulfils the 'gap-free' $\rho_m \sim T^2$ dependence (Fig. 5).

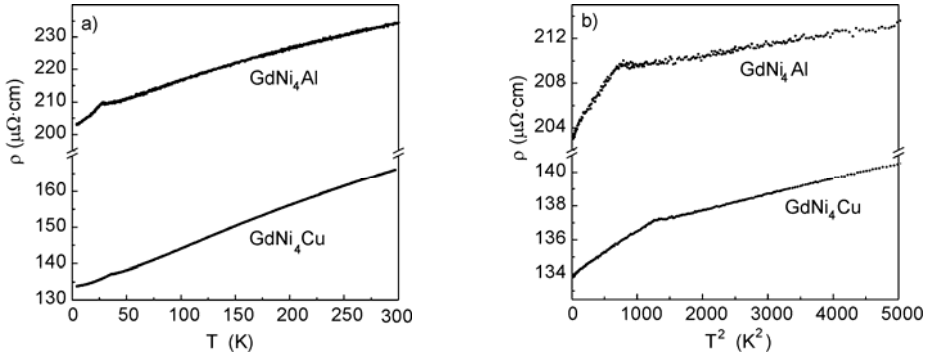

Fig. 5. (a) The $\rho(T)$ curves of the $GdNi_4X$ (X = Al, Cu) compounds as a function of: a) T, b) T^2; the curve b) reflects the scattering on magnons below T_C and the spin fluctuations in a narrow range above the ordering temperature

For all the magnetic RNi_4X compounds the temperature dependence of resistivity in the paramagnetic region – but near T_C – is also quadratic, which is typical of the spin fluctuations ρ_{sf}.

Finally, one can notice that the RNi_4Al, RNi_4Cu and RNi_4B compounds differ dramatically in the value of the residual resistivity ρ_0. For RNi_4Al it is several times higher than for RNi_4Cu but the latter one still has ρ_0 twice the values of the previously studied RNi_4B [1]. It can be explained by the increased disorder in RNi_4Al and RNi_4Cu ($CaCu_5$-type of structure) characterized by a random occupation of the 3g site by the Ni and Al(Cu) atoms [4]. A similar tendency occurs for the spin-disorder resistivity defined as a difference between $\rho_{sf}(T \to 0)$ and ρ_0. The difference in the transport properties of the isostructural RNi_4Al and RNi_4Cu compounds stems probably from the presence of the Cu(3d) states in the valence band region.

3. Conclusions

Based on the Bloch–Grüneisen formula the phonon contribution to $\rho(T)$ has been determined for YNi_4X compounds with the Debye temperatures equal to 204 K, 235 K and 240 K for X = Al, Cu and B, respectively. These values appeared to reflect well the differences in the Y–Y separation in the direction of the hexagonal axis. For the ferromagnetic rare earths the energy gap in the magnon spectrum has been estimated, with the largest value in the case of the $SmNi_4X$ compounds. The $\rho(T)$ dependences of the $CeNi_4X$ compounds show a shallow minimum at low temperatures, which has been ascribed to the Kondo impurity-like effect.

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