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Physical properties of the new ferromagnetic Kondo system $CeAg_{1-x}Ni_xSb_2$

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Structural, magnetic and transport properties of $CeAg_{1-x}Ni_xSb_2$ compounds are investigated on polycrystalline samples. The experimental results show the formation of ferromagnetic compounds with sharp transitions decreasing from 9.5 K in the 100% Ag compound to 8.1 K for 33% Ni substitution, 7 K for 67% Ni substitution to 6 K for the 100% Ni compound. The characteristic temperatures, considered as the temperature of the resistivity maximum T_0 increases from 26 K to 44 K, 54 K and 65 K, respectively. These results indicate Ce(AgNi)Sb2 compounds as a ferromagnetic Kondo system.

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1. Introduction

In the framework of our systematic scan for strongly correlated electron systems among cerium containing alloys we have extended our interest to the structural, magnetic and transport properties of $CeAg_{1-x}Ni_xSb_2$ compounds.

$CeTSb_2$ (T = Ag, Ni) or more generally RTX_2 (R= rare earth) crystallize in the tetragonal $ZrCuSi_2$ - type structure (space group P4/nmm). $CeTSb_2$ structure can be understood from the stacking arrangement of CeSb-T-CeSb-Sb layers [1]. As a result of this arrangement, anisotropic properties may be expected and indeed, e.g. $CeAgSb_2$ [2–4] and $CeNiSb_2$ [5] are exhibiting such strong anisotropy in magnetic and transport properties as demonstrated on single crystal samples.

$CeAgSb_2$ and CeNiSb2 are both moderate heavy fermion ferromagnets with an electronic specific heat coefficient $\gamma \approx 50 mJ/K^2$ mol [3,5] and exhibiting typical Kondo behavior [6,7]. While $CeNiSb_2$ is a ferromagnet with magnetic moments aligned in the basal plane and saturation moment of about 0.7 μ_B/Ce [5], the magnetic behavior of $CeAgSb_2$ is more controversial. Several different groups have investigated the magnetic properties of $CeAgSb_2$ with conflicting results for the interpretation of the magnetic ground state [3,5-9]. For instance, Muro *et al* [7] suggested a ferrimagnetic ground state in polycrystalline samples with a spin-flip field of about 1.3 T. However, this result disagrees with muon spin rotation (μSR) measurements where spectra in both the ordered state and paramagnetic state indicate a single crystallographic and magnetic muon state [9].

In this paper, based on structural, magnetic and transport properties, we describe the $CeAg_{1-x}Ni_xSb_2$ as a ferromagnetic Kondo system.

2. Experimental

$CeAg_{1-x}Ni_xSb_2$ compounds (x = 0, 0.33, 0.67 and 1) have been prepared by RF melting in Ar atmosphere. In order to compensate the Sb loss during preparation we have started with an additional 15% of Sb. The compounds have been investigated by XRD at room temperature, magnetometry using a SQUID magnetometer up to 6 T magnetic field and down to 2 K temperature and magneto transport up to 12 T and down to 300 mK. All compounds were found to crystallize in the tetragonal ZrCuSiAs type structure with the lattice parameter *a* slightly increasing when Ag is substituted by Ni and the lattice constant *c* decreasing, respectively.

3. Results and discussion

Previously results of specific heat, magnetization, and electrical resistivity for all samples $CeAg_{1-x}Ni_xSb_2$ [10] indicate a magnetic order at low temperatures for all compounds. Since single crystal samples of $CeAgSb_2$ [3] and $CeNiSb_2$ [5] have already been characterized as ferromagnetic compounds, we focus on the mixed compounds.

XRD patterns from the $CeAg_{1-x}Ni_xSb_2$ compounds were collected at room temperature on powder samples with grain dimensions below 20 μm. $CeAgSb_2$ was found to be a monophasic sample as shown in figure 1 with an almost stoichiometric composition. The good quality of the sample is further confirmed by the low residual resistivity and the presence of a sharp transition in the temperature dependence of the static susceptibility in low magnetic field.

The XRD patterns of Ag-Ni samples and the $CeNiSb_2$ sample show also smaller amount of impurity phases, with an estimated cumulated amount of about 3.7%. Unfortunately we were not yet able to identify these phases and elemental analysis work is now in progress to solve this problem. However, specific heat measurements as well as low temperature magnetic and transport properties investigations showed no other transitions besides the magnetic transition associated to the main 1:1:2 phase, at least down to 0.3 K. Also the magnetic ordering temperature of $CeNiSb_2$ is 6 K, the same value obtained on single crystals [5]. Thus we concluded that the impurity phases might be associated with the incomplete formation of structure in the intergrain regions or on the grain borders. Preliminary results of the XRD spectra refinement are listed in the Table I.

Table 1. Structural and magnetic parameters of Ce(Ag,Ni)Sb_2 compounds.

compound	a (Å)	c (Å)	c/a	refined composition	T_C [K]
$CeAgSb_2$	4.366(5)	10.705(3)	2.45	$CeAgSb_{\sim 2}$	9.5
$CeAg_{0.67}Ni_{0.33}Sb_2$	4.381(8)	10.659(1)	2.43	$CeAg_{0.64}Ni_{0.19}Sb_2$	8.1
$CeAg_{0.33}Ni_{0.67}Sb_2$	4.407(1)	10.390(2)	2.36	$CeAg_{0.32}Ni_{0.54}Sb_2$	7
$CeNiSb_2$	4.410(5)	9.765(8)	2.21	$CeNi_{0.82}Sb_2$	6

As a general conclusion, the results derived from XRD data show that the Ni-containing compounds have a Ni deficit while the Ag content remains nearly stoichiometric. The *a* lattice parameter increases with the Ni content while the *c* parameter decreases, leading to an overall decrease of the structural anisotropy of about 10% from $CeAgSb_2$ to $CeNiSb_2$.

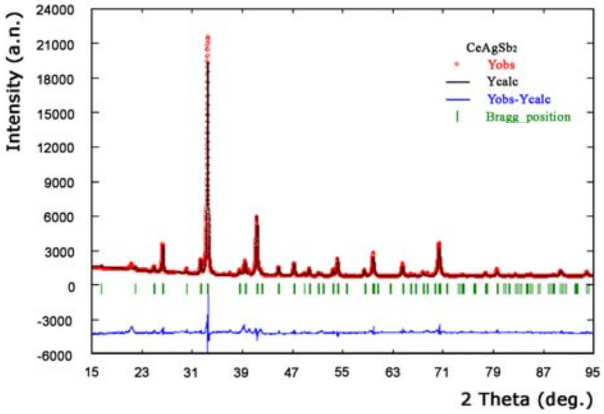

Fig. 1. XRD data analysis for polycrystalline $CeAgSb_2$ compound.

Isotherm magnetizations recorded down to 3 K are shown in Fig. 2 (left) for the Ce(Ag,Ni)Sb_2 compounds. For all compounds, the field dependencies recorded at lowest temperatures indicate a ferromagnetic behavior. The saturation value of the magnetization decreases with increasing temperature for all compounds.

Low field temperature dependencies of the magnetization indicate ferromagnetic transitions from 9.5 K in $CeAgSb_2$ to 6 K in CeNiSb2. The values derived for all compounds are listed in the last column of Table I. These values are in perfect agreement with those obtained from specific heat and resistivity measurements. The FM order is further confirmed by the presence of spontaneous magnetization inferred from Arrott plots obtained in the vicinity of the ordering temperatures. In Figure 2 (right) the corresponding Arrott plots are depicted. Similar points color and shape are used for curves where no spontaneous magnetization can be observed.

The temperature dependencies of electrical resistivity can be used to trace the evolution of the system (see Fig. 3). Below the magnetic ordering temperature the resistivity drops due to the suppression of the spin disorder scattering contribution to the electrical resistivity. The magnitude of the resistivity drop can be related on one side to the value of the residual resistivity and on the other side to the anisotropy effect. Above the ordering temperature the resistivity slope exhibit a broad maximum which can be ascribed to the Kondo effect in the presence of a crystalline electric field (CEF), in the scope of Coqblin-Schrieffer model. In this case, the characteristic temperatures considered as the temperature of the resistivity maximum T_0 increases from 26 K to 44 K, 54 K and 65 K, for x=0, 0.33, 0.67 and 1 respectively.

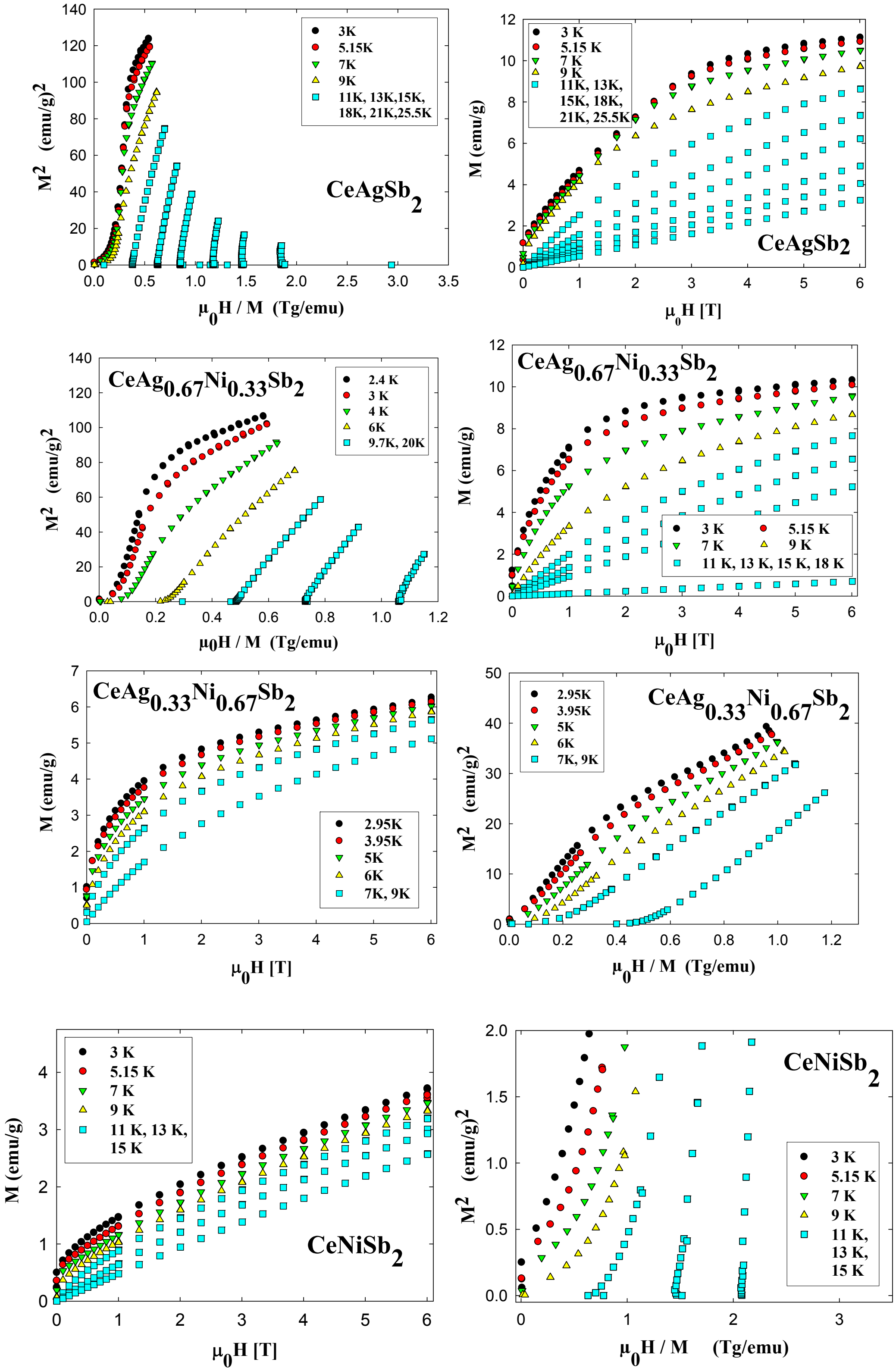

Fig. 2. Magnetization isotherms (left) and the corresponding Arrott plots (right) for $CeAg_{1-x}Ni_xSb_2$ compounds.

A survey of data plotted in Fig. 3 shows that with increasing fields the characteristic temperature T_0 increases as expected from Ce-based Kondo systems, while the magnetic ordering temperature cannot be observed anymore due to the strong broadening of the maximum. However, excepting the Ni sample and only at 12 T, a non-Fermi liquid behavior of the resistivity was not detected. This means that the ordering temperature remains well above 0 K for ambient pressures even at the maximum applied field.

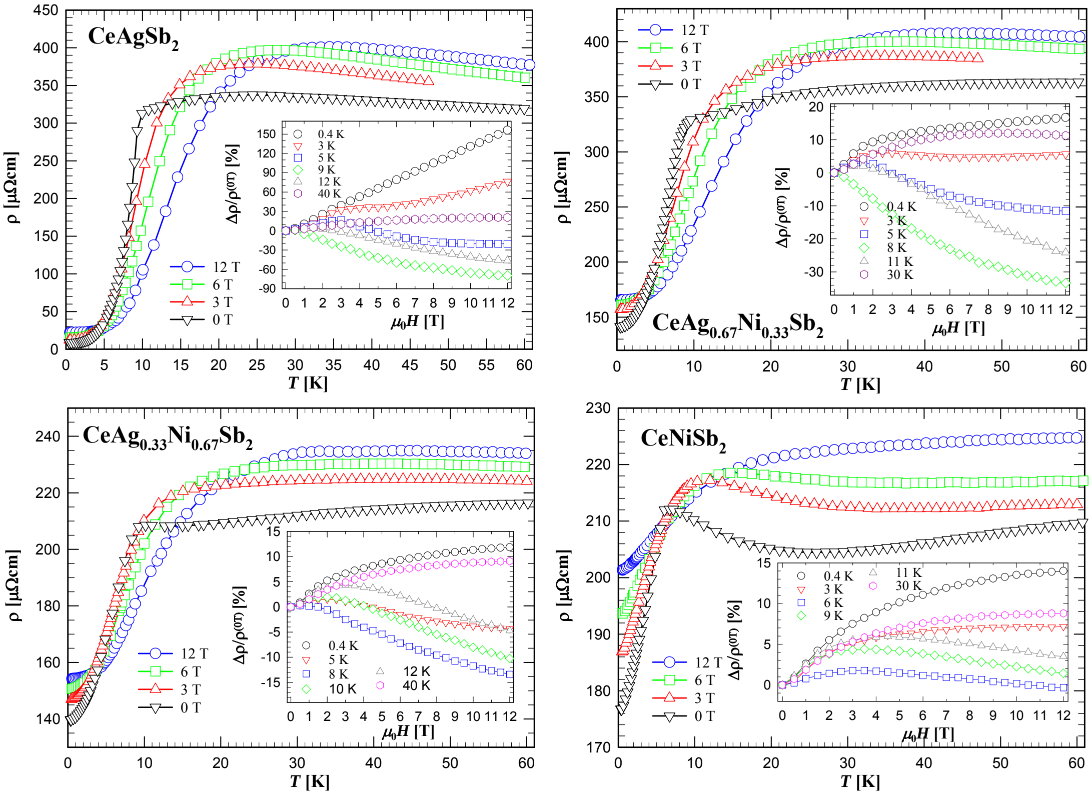

Fig. 3. Electrical resistivity as function of temperature for different applied magnetic fields in the Ce(Ag,Ni)Sb$_2$ system (main graphics) and the corresponding magnetorezistivity isotherms (insets).

The magnetoresistivity isotherms for the Ce(Ag,Ni)Sb$_2$ compounds are plotted in the graphics insets in Figure 3. A striking feature which can be immediately observed is the huge magnetoresistivity of CeAgSb2 which reaches about 150% at lowest temperature 0.4 K. A similar behavior was also observed in reference [12] on CeAgSb$_2$ under pressure and was ascribed to the suppression of magnetic order at high pressures. An external magnetic field might also be responsible for suppression of magnetic order, but in this particular case a critical behavior is expected to occur at much higher field values. Most likely, here the huge magnetic resistivity can be the result of cumulative effects as the rotation of magnetic moments with different orientations in a polycrystalline sample and the peculiar magnetic behavior of this compound which in spite of a ferromagnetic alignment along *c*-axis has also a large magnetization along the *a*-axis. This unusual magnetic behavior was explained in terms of the CEF level scheme with an anisotropic exchange interaction [13]. A similar behavior is also observed in the other Ce(Ag,Ni)Sb$_2$ compounds, however with maximal magnetoresitivities around only to 20% (compare Figure 3, insets). As the temperature increases, the magnetoresistivity decreases and becomes negative, with a minimum around the ordering temperature of the compound. This minimum value also decreases with increasing Ni content, from about 60% in CeAgSb$_2$ to only 1% in CeNiSb$_2$. Above the ordering temperature the magnetoresistivity tends to the usual paramagnetic and positive shape.

A crude analysis of the experimental temperature dependencies of the resistivityt in magnetic field was performed taking into account the electron magnon scattering using the same model used for CeAgSb$_2$ single crystals by Jobiliong et al [11]. Fit results using the contribution due to electron ferromagnetic magnons scattering, i.e. $\rho = \rho_0 + BT\Delta(1+2T/\Delta)e^{-\Delta/T}$, where ρ_0 is the residual resistivity and Δ is the spine-wave gap suggests that the both *B* and Δ are decreasing with increasing field. Test fits were also performed using assuming an antiferromagnetic order, but they were limited to much lower temperatures. The corresponding Δ^{AFM} values were much smaller than Δ^{FM}, in agreement with the results obtained on single crystals [11].

4. Summary

Concluding, we have presented the structural, magnetic and transport properties of $CeAg_{1-x}Ni_xSb_2$ system. The $CeAg_{1-x}Ni_xSb_2$ compounds were found to crystallize in the tetragonal ZrCuSiAs type structure. Magnetic, specific heat and transport properties investigations indicate a ferromagnetic Kondo behavior for all the compounds.

Acknowledgements

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