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Study of Magnetic Contribution to the Heat Capacity of $YbCu_4Ni$

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An investigation of the magnetic heat capacity of the heavy-fermion compound $YbCu_4Ni$ up to 0.4 K is presented. The novel compound $LuCu_4Ni$, isotypic with $YbCu_4Ni$ has been synthesized and characterized, and its heat capacity was measured in order to subtract the lattice contribution from the previously measured heat capacity data of $YbCu_4Ni$.

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1. Introduction

Strong correlation between electrons, due to hybridization of f-electrons and conduction electrons, can cause a number of outstanding low temperature features. Among the rare earths, a large number of these phenomena is found for Ce- and Yb-based compounds [1–3]. The interest in this topic was triggered by the investigation on the heavy fermions $YbCu_4T$ (T = Ag, Au), which crystallize in an ordered derivative of the $AuBe_5$ structure type (cubic $MgCu_4Sn$ type) [4–6]. It was found that $YbCu_4Au$ orders antiferromagnetically below 1 K [7], whereas $YbCu_4Ag$ has a nonmagnetic ground state [7].

Recently the novel compound $YbCu_4Ni$ has been studied [8]. It was discovered that this compound is a new heavy fermion (HF) member of the series of $YbCu_4M$ (M = metal) and it crystallizes in the cubic $MgCu_4Sn$ type structure. The measurement of heat capacity revealed a possible magnetic ordering below 0.5 K, which was also confirmed by susceptibility measurements. In this paper we present the results of the investigation of the magnetic contribution to the heat capacity of $YbCu_4Ni$ till 0.4 K. The novel compound $LuCu_4Ni$, isotypic with $YbCu_4Ni$, has been prepared for the first time in this work, and we present its temperature dependence of heat capacity in order to determine the magnetic contribution and the magnetic entropy of $YbCu_4Ni$. The measurements have been done in the temperature range 0.4–300 K and in applied magnetic fields up to 9 T.

2. Experimental

We have used the heat capacity data reported in our previous paper [8]. The details on preparation of the polycrystalline sample of $YbCu_4Ni$ have been already reported [8]. The preparation of new polycrystalline sample of $LuCu_4Ni$ started by weighting the stoichiometric amount of elements with the following nominal purity: La (99.9 pct mass), Cu (99.999 pct mass), and Ni (99.995 pct mass). Then the elements were enclosed in a small tantalum crucible and sealed by arc welding under pure argon. The sample has been then melted in an induction furnace under a stream of pure argon. To ensure homogeneity during the melting, the crucible was continuously shaking. The sample was then annealed in a resistance furnace at 700 °C for two weeks and finally quenched in cold water. The crystal structure was examined by X-ray diffraction (XRD) using a Philips diffractometer (Cu K_α radiation). The Rietveld matrix full profile structure refinement was carried out using the program FULLPROF [9].

The results of the Rietveld refinement carried out on $LuCu_4Ni$ are presented in Table. The starting atomic position parameters were taken from the crystallographic data of the cubic $MgCu_4Sn$ structure type and the reliability factors obtained are $R_F = 0.065$ and $R_B = 0.0628$. The data in Table show a very good agreement with the model of the cubic $MgCu_4Sn$ structure type, with a full ordered distribution of the atoms at the sites of the structure. At first, the atomic occupations were allowed to vary, showing very small differences from unity. Thus, their values were fixed to unity (100% in Table).

TABLE

Structural parameters of $LuCu_4Ni$ refined from X-ray data in the space group F-43m. "Occ." = occupation number; residual values: $R_F = 0.065$, $R_B = 0.0628$, $R_{WP} = 0.0532$; * = fixed.

Atom	Site	x	y	z	Occ. [%]
Lu	4c	0.25	0.25	0.25	100.0*
Cu	16e	0.6258(1)	0.6258(1)	0.6258(1)	100.0*
Ni	4c	0	0	0	100.0*

Figure 1 shows the Rietveld plot obtained for the best agreement between calculated and observed profiles. The refined lattice parameter is $a = 6.9326(2)$ Å.

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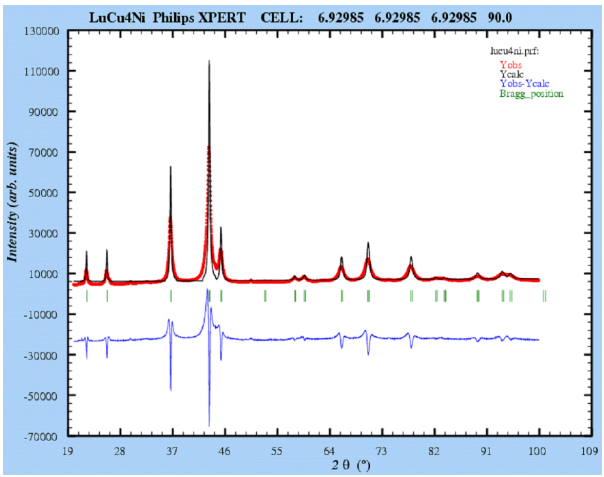

Fig. 1. X-ray diffraction powder pattern of $LuCu_4Ni$ compared with the calculated diffraction diagram. The experimental data are shown by symbols, while the line through the data represents the results of the Rietveld refinement. The lower curve is the difference curve. The ticks indicate the 2θ values of the Bragg peaks.

Heat capacity measurements were performed by PPMS commercial device (Quantum Design) in the temperature range 0.4–300 K and in an applied magnetic field up to 9 T. The heat capacity was measured using the two-τ model of the relaxation method.

3. Results and discussion

In Fig. 2 the low temperature part (0.4–20 K) of the heat capacity $C(T)$ of polycrystalline $LuCu_4Ni$ measured for various values of applied magnetic fields up to 9 T, is shown. No influence of a magnetic field in the whole measured temperature range, and up to 9 T is evidenced in $C(T)$, which points to a negligible magnetic contribution to the measured heat capacity. This behaviour is characteristic of ordinary metals with no sign of a phase transition. In order to confirm it we determined the electronic coefficient γ by analysing the dependence of C/T as a function of T^2. We have obtained $\gamma = 9.5$ mJ/(mol K^2). The estimated Debye temperature is $\theta_D = 305$ K. Thus this compound is a good non-magnetic phonon reference for $YbCu_4Ni$ and therefore it was used to determine the magnetic contribution $C_{mag}(T)$ to the heat capacity of $YbCu_4Ni$ previously measured in [8].

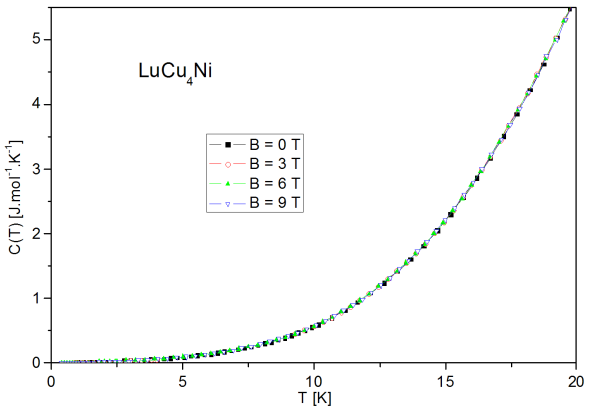

Fig. 2. Low temperature detail of the temperature dependence of the heat capacity $C(T)$ of $LuCu_4Ni$ as a function of an applied magnetic fields up to 9 T.

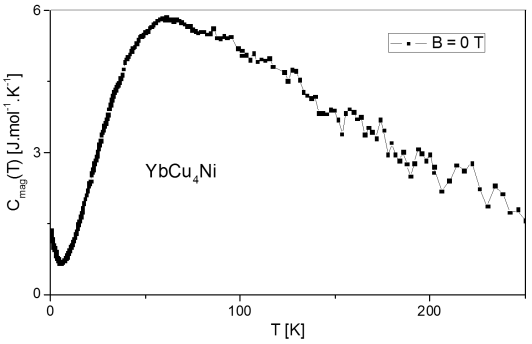

Fig. 3. Temperature dependence of the magnetic contribution $C_{mag}(T)$ to the heat capacity of $YbCu_4Ni$ at zero magnetic field.

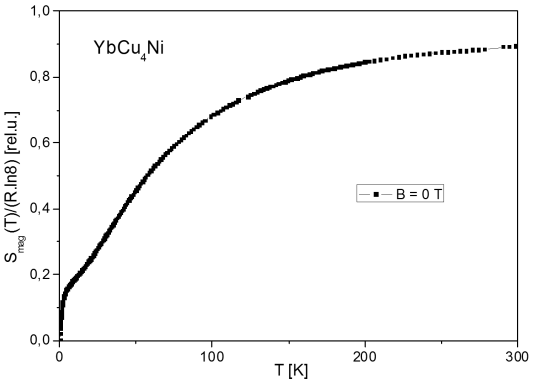

Fig. 4. Temperature dependence of the magnetic entropy $S_{mag}(T)$ of $YbCu_4Ni$ at zero magnetic field.

In Fig. 3 the temperature dependence of the magnetic heat capacity $C_{mag}(T)$ is shown. For temperatures $T < 5$ K an upturn is present, which may be due to a possible magnetic transition below 0.5 K. Moreover, a large broad maximum is observed at about 60 K. This maximum is a Schottky anomaly connected with the crystal electric field (CEF) effect on the Yb ion in the cubic structure.

In Fig. 4 the plot of the magnetic entropy ratio $S(T)/R\ln 8$ determined from $C_{mag}(T)$ is shown, where R is the gas constant and number 8 is the degeneracy $(2J+1)$ of the Hund ground state with $J = 7/2$ for Yb^{3+}. The entropy increases with increasing temperature, reaching at 300 K the value of 15.44 J K^{-1} mol^{-1}, which is still below the theoretical value $R\ln 8 =$

17.28 J K^{-1} mol^{-1}. A rather low magnetic entropy was found despite the fact that the ground state is a doublet. An entropy value $S = R \ln 2$, associated with such a ground state, is achieved at about $T = 32$ K, i.e. well above the expected magnetic transition temperature. This behaviour is similar to those reported for other $YbCu_4X$ compounds [7].

4. Conclusions

In conclusion, a novel $LuCu_4Ni$ compound, crystallizing in the cubic structure of $MgCu_4Sn$ type, has been prepared. The heat capacity of $LuCu_4Ni$ in the temperature range 0.4–300 K and in applied magnetic fields up to 9 T has been studied. We have observed no influence of the magnetic field on $C(T)$. The estimated Debye temperature of 305 K and the γ value of 9.5 mJ/(mol K^2) indicated $LuCu_4Ni$ as a suitable candidate in order to determine the magnetic heat capacity of the heavy fermion $YbCu_4Ni$ (with an isotypic structure). We determined the magnetic entropy which value, at room temperature, is still below the theoretical value.

Acknowledgments

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