Proceedings of the CSMAG'07 Conference, Košice, July 9–12, 2007

Lattice Heat Capacity in RTAl (R = Y, Lu; T = Ni, Cu, Pd) Compounds

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We present analysis of the specific heat of the RTAl (R = Y, Lu; T = Cu, Ni, Pd) compounds. We focus on the lattice contribution and analyze the dependence of all the characteristic parameters in the Debye and Einstein models on the atomic masses and interatomic distances.

PACS numbers: 65.40.Ba, 75.40.Cx

1. Introduction

Among the ternary RTAl compounds (R = rare earth, T = transition metal) those with T = Ni, Cu or Pd crystallize in the hexagonal ZrNiAl-type structure (P-62m group). All the three series show interesting magnetic behavior. Important contribution to the understanding of the electronic properties of these materials is obtained from the specific-heat data. To analyze correctly all the contributions, including the magnetic one, one has to estimate the contribution due to lattice vibrations, $C_{\rm ph}$. This phonon contribution is the dominating one above $\approx$ 20 K, depending also on the intrinsic properties of the material. It is therefore very important to estimate the phonon part as close to the reality as possible. In this paper, we present the specific heat of all the non-magnetic RTAl compounds with the ZrNiAl-type structure. We analyze the dependence of all the characteristic parameters on the atomic masses and interatomic distances.

2. Experimental

The polycrystalline samples were prepared by arc-melting in mono-arc furnace under protection of argon atmosphere. The samples were analyzed by X-ray powder diffraction at room temperature with a conventional diffractome-

ter (Siemens) working with the Bragg–Bretano geometry. The specific heat was measured utilizing the relaxation method using the Quantum Design PPMS system on samples with the weight of 20–30 mg in the temperature range between 2 K and 300 K.

3. Results and discussion

There are six RTAl compounds with the ZrNiAl-type structure with the non-magnetic rare earth: YTAl and LuTAl with T = Ni, Cu, or Pd. The X-ray analysis showed all the studied compounds to be single phase with the ZrNiAl-type hexagonal structure. The lattice parameters are listed in Table. Compounds with La unfortunately do not form this crystal structure [2]. The specific heat of the

TABLE

Parameters characterizing the crystal structure and specific heat. The parameters θ_{E1} and θ_{E2} describe 2 and 4 branches, respectively. The lattice parameters for YCuAl and LuCuAl are taken from Ref. [1].

	a [pm]	c [pm]	γ [mJ/(mol K)]	θ_D [K]	θ_{E1} [K]	θ_{E2} [K]
YNiAl	703.2(2)	383.8(1)	8.1(0.5)	214(3)	186(10)	360(30)
YCuAl	703.5	403.5	5.7(0.4)	238(3)	156(10)	324(30)
YPdAl	722.6(2)	393.8(1)	5.9(0.5)	177(4)	174(10)	323(30)
LuNiAl	694.3(2)	375.7(1)	7.2(0.5)	174(3)	175(10)	353(30)
LuCuAl	691.2	399.7	4.7(0.5)	205(4)	140(10)	335(30)
LuPdAl	715.3(2)	385.1(1)	5.7(0.6)	150(3)	159(10)	311(30)

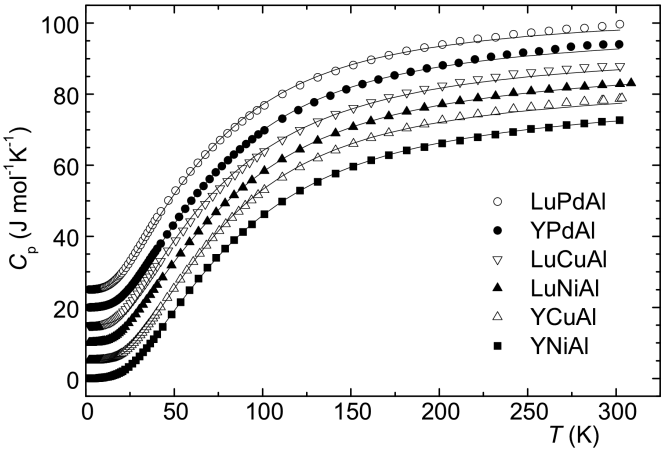

Fig. 1. The experimental specific heat of the studied compounds; solid lines represent the curves calculated using the parameters given in Table. Data for individual compounds are shifted by 5 J mol^{-1} K^{-1}.

studied non-magnetic compounds, shown in Fig. 1, consists only of the electronic, $C_{\rm el} = \gamma T$, and the phonon, $C_{\rm ph}$, contributions. The phonon spectrum in the case of studied compounds consists of 3 acoustic and 6 optic branches. In order to obtain a satisfactory expression for $C_{\rm ph}$, we applied the Debye model to the 3 acoustic branches, and the Einstein model to the 6 optic branches

$$C_{\rm ph} = 9R\left(\frac{T}{\theta_{\rm D}}\right)^3 \int_0^{\frac{\theta_{\rm D}}{T}} \frac{x^4 {\rm e}^x}{\left({\rm e}^x - 1\right)^2} {\rm d}x + R\sum_{i=1}^{6}\left(\frac{\theta_{{\rm E}i}}{T}\right)^2 \frac{{\rm e}^{\theta_{{\rm E}i}/T}}{\left({\rm e}^{\theta_{{\rm E}i}/T} - 1\right)^2}, \qquad (1)$$

where the first term describes the three acoustic branches and the second term is due to the optic branches, $\theta_{\rm D}$ and $\theta_{{\rm E}i}$ are characteristic Debye and Einstein temperatures, respectively, R is the gas constant.

At sufficiently low temperatures ($T \ll \theta_{\rm D}$), the contribution of optic branches to the C_p is negligible and the Debye model simplifies to βT^3. The total specific heat can be then written as

$$C_p = \gamma T + \beta T^3, \qquad (2)$$

where β is related to $\theta_{\rm D}$. As a first step, we have used this equation to fit our low-temperature data (up to 8 or 10 K) and to determine the γ and $\theta_{\rm D}$ parameters (see Fig. 2). In a second step, we fit Eq. (1) to data from the whole measured

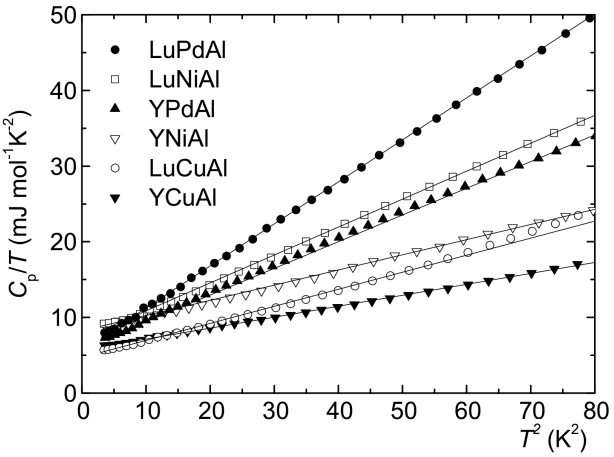

Fig. 2. Specific heat of the studied compounds at low temperatures; solid lines represent the fit to Eq. (2).

range with fixed γ and $\theta_{\rm D}$. We obtain the $\theta_{{\rm E}i}$ values as a result. The sensitivity of such fit (considering also experimental errors of our data, $\approx$ 1–4%) to individual $\theta_{{\rm E}i}$ is rather low and we can obtain several very different schemes giving similar agreement with the measured data. It is thus reasonable to reduce the number of different $\theta_{{\rm E}i}$. On the other hand, the data cannot be satisfactory described by a single $\theta_{\rm E}$. Finally, we used two parameters $\theta_{\rm E1}$ and $\theta_{\rm E2}$ describing two and four optic branches, respectively. Results are summarized in Table. We are aware that

it is only a model far from reality of complex phonon spectra which gives only basic characteristics of the lattice dynamics.

Let us now discuss the results. The electronic contribution is small as expected. The γ is higher for the RNiAl than for RCuAl, which is consistent with higher density of state at E_{F} for Ni. The θ_{D} and θ_{E} values depend generally on the atomic masses and bond stiffness. The mass dependence can qualitatively account for lower θ_{D} in Lu compounds compared to the Y counterparts and also for low θ_{D} in the Pd compounds. On the other hand, the mass difference cannot account for rather different θ_{D} in Ni and Cu compounds. The interatomic distances (and thus the bond stiffness) play a crucial role here. The present analysis did not reveal any clear trends for the optic phonons, except that θ_{E1} seem to be, at least qualitatively when comparing mutually Y or Lu compounds, inversely proportional to the c lattice parameter.

We can conclude that the lattice vibrations in RTAl compounds are equally influenced by both factors, the atomic masses and the interatomic distances. Any exact quantitative predictions for RTAl with other, magnetic rare earth is therefore practically impossible, especially when the La analogues cannot be included in the analysis. Considering the atomic masses and the lattice parameters in the whole RTAl series, we suggest to use directly the LuTAl measured data as an approximation for the phonon part in RTAl for R = Tb to Tm. The estimation for compounds with light rare earth should be treated individually with higher uncertainty.

Acknowledgments

The work was supported by the Grant Agency of the Czech Republic under the grant No. 106/05/0393 and is a part of the research program MSM 0021620834 financed by the Ministry of Education of the Czech Republic.

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