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Specific Heat of $YbNi_4Si$ Compound

M. Falkowski *, A. Kowalczyk and T. Toliński

Institute of Molecular Physics, Polish Academy of Sciences
Smoluchowskiego 17, 60-179 Poznań Poland

The specific heat of $YbNi_4Si$ has been analyzed considering the electronic contribution and the lattice contributions in frames of the Debye model. Based on the specific heat measurements, the electronic specific heat coefficient $\gamma = 25$ mJ mol^{-1} K^{-2} and the Debye temperature $\theta_D = 320$ K were derived. This small value shows that $YbNi_4Si$ cannot be classified as a heavy fermion system. These studies are completed by magnetic susceptibility, X-ray photoemission spectroscopy, and electrical resistivity.

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1. Introduction

The intermediate valence compounds pose one of the most challenging problems of strongly correlated electron systems. Different ingredients contribute to the complexity of these fascinating systems: the presence of strong Kondo interactions, the level structure of the crystal electric field split f-orbitals, a different hybridization between each level and the conduction band, and the eventual coherence effects and magnetic interactions introduced by the periodicity of the Kondo lattice.

A great interest in the Yb-based compounds comes from the fact that Yb can exhibit two valence states related to the nonmagnetic $4f^{14}$ (Yb^{2+}) and magnetic $4f^{13}$ (Yb^{3+}) electronic configurations. Due to this feature Yb is often treated as a hole counterpart of Ce, which also shows two close in energy, electronic configurations: the magnetic $4f^1(Ce^{3+})$ and the nonmagnetic $4f^0(Ce^{4+})$.

In our paper we analyze the heat capacity data of $YbNi_4Si$. To verify fully the magnetic contribution due to the Yb f states the nonmagnetic reference compound YNi_4Si is also measured.

2. Experimental

The sample preparation was described in detail in Ref. [1]. The room temperature powder X-ray diffraction pattern revealed that the studied $YbNi_4Si$ com-

*corresponding author; e-mail: falkowski@ifmpan.poznan.pl

pound was single-phase. Heat capacity measurements were performed by PPMS commercial device (Quantum Design) in the temperature range of 2–300 K by the relaxation method using the two-tau model.

3. Results and discussion

$YbNi_4Si$ is paramagnetic and its magnetic susceptibility follows the Curie–Weiss law $\chi(T) = C/(T-\theta)$ with $\mu_{\rm eff} = 4.15\mu_{\rm B}$/f.u. and the paramagnetic Curie temperature $\theta \approx 0$ K [1]. This effective magnetic moment is slightly reduced with respect to the free Yb^{3+} ion ($4.54\mu_{\rm B}$) and suggests that the valence of the ytterbium ions in $YbNi_4Si$ is close to 3+. The Yb^{2+} and Yb^{3+} peaks observed by X-ray photoemission spectroscopy (XPS) in the valence band region confirm the domination of the Yb^{3+} valence state [1] (Fig. 1).

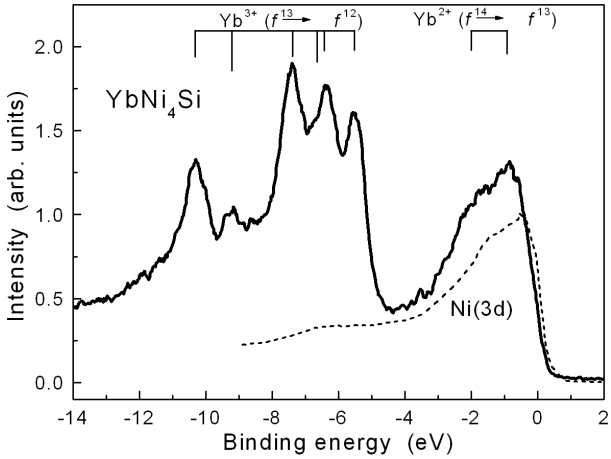

Fig. 1. Valence band region of the $YbNi_4Si$ compound measured by X-ray photoemission spectroscopy.

The temperature dependence of the electrical resistivity $\rho(T)$ shows the standard Fermi-liquid behavior with T^2 dependence up to about 60 K.

The total specific heat consists of electronic and phonon contributions. The electronic specific heat coefficient γ provides information concerning the conduction band density of states at the Fermi level. Figure 2 shows the temperature dependence of the specific heat for YNi_4Si and $YbNi_4Si$ compounds.

YNi_4Si is nonmagnetic; therefore, $C_p(T)$ can be described by the standard formula:

$$C_p(T) = \gamma T + 9NR\left(\frac{T}{\theta_{\rm D}}\right)^3 \int_0^{\theta_{\rm D}/T} \frac{x^4 \exp(x)\mathrm{d}x}{[\exp(x)-1]^2} \quad , \tag{1}$$

where the first and the second term correspond to the electronic and the phonon contribution, respectively. $N = 6$ is the number of the atoms in the formula unit and $x = h\omega/k_{\rm B}T$. From the fit to the experimental points (Fig. 2) we get

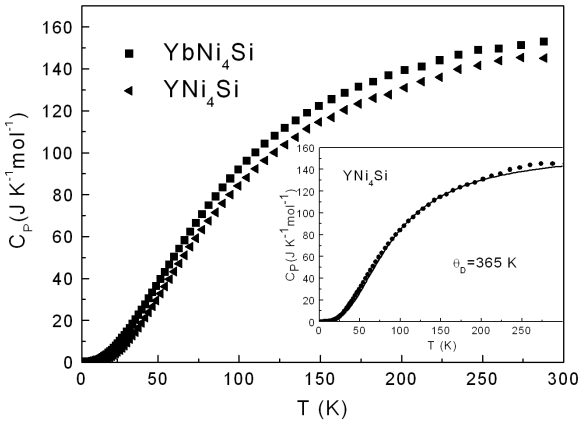

Fig. 2. Temperature dependence of the specific heat of $YbNi_4Si$ and YNi_4Si. (Inset) Specific heat of the YNi_4Si compound fitted with Eq. (1) (solid line).

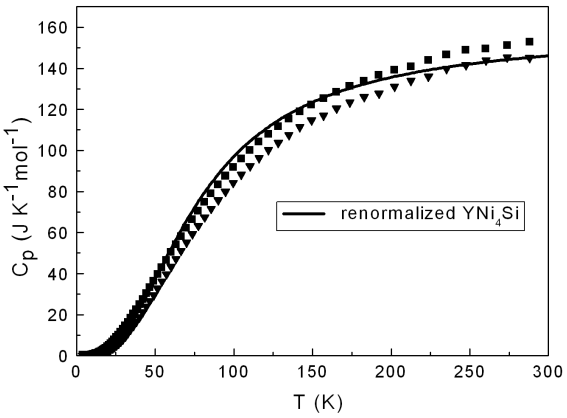

Fig. 3. The specific heat of $YbNi_4Si$ (squares) and YNi_4Si (triangles). Solid line shows the YNi_4Si data after renormalization of the mass in respect to the Yb-based compound.

the Debye temperature $\theta_D = 365$ K and the electronic specific heat coefficient $\gamma = 13$ mJ mol^{-1}K^{-2}. A similar analysis has been carried out for $YbNi_4Si$ yielding $\theta_D = 320$ K and $\gamma = 25$ mJ mol^{-1}K^{-2}, respectively. However, if a significant magnetic contribution is expected for the Yb-based compound then the simple Debye model does not describe the lattice specific heat accurately enough. In the absence of a measurable magnetic part the Debye temperature of $YbNi_4Si$ should be easilly obtained from the data for YNi_4Si by considering a correction to account for the difference between the molar masses of the $YbNi_4Si$ and YNi_4Si compounds. The renormalization ratio [2] is expressed as:

$$\frac{\theta_{\mathrm{D}}(R_mX_nY_p)}{\theta_{\mathrm{D}}(R'_mX_nY_p)} = \left(\frac{m(M_{R'})^{3/2} + n(M_X)^{3/2} + p(M_Y)^{3/2}}{m(M_R)^{3/2} + n(M_X)^{3/2} + p(M_Y)^{3/2}}\right)^{1/3}, \tag{2}$$

where M_R, M_X and M_Y are the molar masses of the R, X, and Y atoms, respectively, θ_{D} is the Debye temperature and can be used to evaluate the lattice contribution of the magnetic compound $R_mX_nY_p$ by rescaling the data for the nonmagnetic $R'_mX_nY_p$. reference system.

Figure 3 presents the specific heat of $YbNi_4Si$ (squares) and YNi_4Si (triangles) together with the renormalized data of YNi_4Si (solid line). The Debye temperatures of $YbNi_4Si$ obtained from the direct fit with Eq. (1) and from the renormalization of YNi_4Si are nearly identical, which indicates that the magnetic contribution to the specific heat of $YbNi_4Si$ is very small, therefore we omit its analysis.

4. Conclusions

It has been found that the $YbNi_4Si$ compound does not order magnetically down to 4 K. Nearly in the whole temperature range studied the magnetic susceptibility follows a Curie law with $\mu_{\mathrm{eff}} = 4.15\mu_{\mathrm{B}}$/f.u. This effective magnetic moment is close to the value expected for the $4f^{13}$ configuration ($4.54\mu_{\mathrm{B}}$). The Yb^{2+} and Yb^{3+} peaks observed by XPS in the valence band region confirm the domination of the Yb^{3+} valence state. The heat capacity measurements have shown that $YbNi_4Si$ is characterized by $\gamma = 25$ mJ mol^{-1}K^{-2} and $\theta_{\mathrm{D}} = 320$ K. A mass correction to the nonmagnetic reference sample YNi_4Si indicates that the magnetic contribution to the specific heat of the Yb-based compound is very small.

References

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[2] M. Bouvier, P. Lethuillier, D. Schmitt, *Phys. Rev. B* **43**, 13137 (1991).