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Thermoelectric Power of $CeNi_4Si$ and $YbNi_4Si$ Compounds

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The thermoelectric power was measured from 4.2 to 300 K for $CeNi_4Si$ and $YbNi_4Si$. The thermoelectric power was analysed in the framework of the phenomenological resonance model. According to the model the dominant contribution to thermopower is caused by scattering between electrons of a broad s-band and a narrow f-band with the Lorentzian shape. The electron–hole analogy is reflected in the thermoelectric power behaviour of the investigated compounds.

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1. Introduction

The intermetallics $CeNi_4Si$ and $YbNi_4Si$ belong to large family of rare-earth RNi_4Si (R = rare earth) compounds, crystallising in the hexagonal $CeCu_5$-type structure (space group $P6/mmm$) [1]. The first compound has been reported to show paramagnetic properties down to 2 K. However, the effective moment deduced from a fit of magnetic data to the Curie–Weiss law is rather low (0.52 μ_B), if one compares to that of the free Ce^{3+} ions (2.54 μ_B). This behaviour was interpreted in terms of intermediate valence of the Ce atoms and may be supported by the X-ray photoemission spectroscopy (XPS) experiments [1]. The latter have shown the f occupancy n_f to be 0.91 and the hybridisation energy $\Delta = 36$ meV between f states and conduction electrons.

Moreover, for $CeNi_4Si$ one finds a minimum in the temperature dependence of the electrical resistivity at 8 K, resembling the behaviour of a single-ion Kondo system. $YbNi_4Si$ seems to be also an intermediate valence compound [1]. Indeed, the XPS experiments have indicated that the majority contribution to the XPS

spectra comes just from the Yb^{3+} ions. The magnetic data consistently with the XPS data show a wide temperature range of the Curie law behaviour with the effective moment (4.15 μ_B) close to that expected for the f^{13} configuration (4.54 μ_B). The information about Ce and Yb compounds mentioned above makes up new support for the concept of electron–hole analogy between Ce and Yb ions. This could be understood since the Yb atoms can appear in two valence states, nonmagnetic (f^{14}) and magnetic (f^{13}). In turn, Ce, which is the electron counterpart of Yb may possess magnetic (f^1) and nonmagnetic (f^0) valence states. Because the thermoelectric power is sensitive to the curvature of the bands at the Fermi level, the comparison of thermoelectric power (TEP) between $CeNi_4Si$ and $YbNi_4Si$ is highly desired. Thus, in the contribution we present experimental data of the thermoelectric power for these compounds and discuss them shortly in the framework of the existing theories.

2. Experimental

TEP was measured in the temperature range 4.2–300 K using the differential method. A temperature gradient of about 1 K was applied along the sample length and was determined using a pair of Au-0.07% Fe/chromel thermocouple. The absolute uncertainty of the measured Seebeck coefficient S is less than 0.5 μV/K.

3. Results

The behaviour of TEP of Ce- and Yb-based intermetallics compounds often differs from those of the ordinary metals and their alloys. Instead of the magnitude of several μV/K, the TEP of compounds with unstable f-electron shells may attain a giant value, i.e., of the order of a hundred μV/K. Furthermore, in addition to the diffusion mechanism that appears as a linear temperature dependence term in the $S(T)$ function, there are other contributions, for instance those, related to the Kondo effect, crystalline electric field or scattering of conduction electrons on a narrow f band. As a result, the DOS structure of these compounds near E_F is rather complicated, and it can lead to the appearance of several extrema in the $S(T)$ curve.

Figure 1a shows the temperature dependence of the thermoelectric power for $CeNi_4Si$. TEP is found to be negative at room temperature. It increases steadily with decreasing temperature until reaches a maximum value of 2.5 μV/K at $T_{max} = 55$ K. Furthermore, the thermopower exhibits a minimum of ≈ -1 μV/K at $T_{min} = 8$ K, followed by a linear dependence $S(T) \sim T$ between 4–7 K. Although the absolute value of TEP of $CeNi_4Si$ is small, it shows a similar structure as many other Ce-based compounds do, for instance, antiferromagnets like CePdSn [2] and $CeAl_2$ [3].

The TEP of $YbNi_4Si$ below 20 K exhibits rather a linear dependence on temperature (Fig. 1b). The linearity of the $S(T)$ curve of $YbNi_4Si$ for $T > 200$ K is doubtlessly related to the diffusion thermopower and the phonon drag certainly

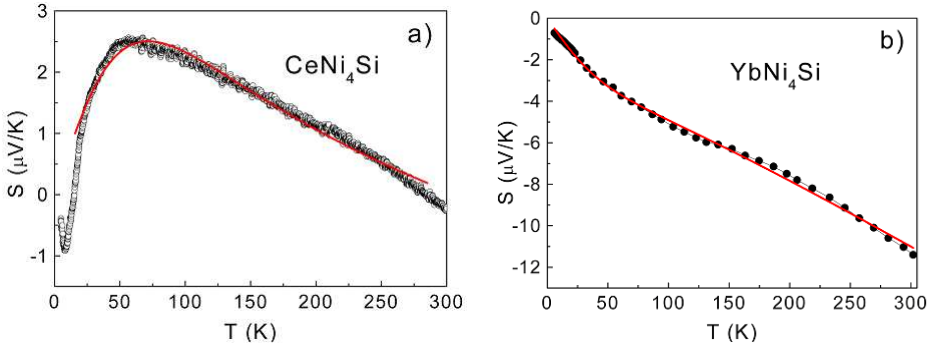

Fig. 1. Temperature dependence of the thermoelectric power of: (a) $CeNi_4Si$, (b) $YbNi_4Si$. The solid lines are fits of the experimental data with the modified Mott's equation.

plays a minor role. Besides, as is typical of Ce- and Yb-based compounds TEP is mostly positive for the former and negative for the later [4]. It is a direct consequence of the electronic structure (Yb being a hole counterpart of Ce). One can notice that TEP in the temperature range 25–175 K deviates from the linear dependence. Since the studied compound is nonmagnetic, we can neglect the magnon drag.

Ce compounds in the valence fluctuation regime exhibit a large maximum in $S(T)$. To explain this observation, Gottwick et al. [5] proposed a phenomenological model based on the two-band conductor model, which was originally proposed for 3D transition metals and alloys. The conduction electrons are assumed to be scattered by a $4f$ quasiparticle band of a Lorentzian form. Modifying Mott's equation, the TEP is expressed as

$$S_B(T) = \frac{AT}{B^2 + T^2} \tag{1}$$

with

$$A = \frac{2\Delta}{|e|} \quad \text{and} \quad B^2 = 3\frac{\Delta^2 + \Gamma^2}{\pi^2 k_{\mathrm{B}}^2}. \tag{2}$$

The parameter $\Delta = E_0 - E_{\mathrm{F}}$ is a measure of the position of the DOS peak in respect of the Fermi level and Γ is the width of the $4f$ band.

The fits presented in Fig. 1a include additionally the linear term due to the diffusion thermopower, i.e., $S(T) = S_B(T) + cT$. The parameters have the values: $\Delta = 0.23$ meV and $\Gamma = 24$ meV, with $c = -0.0045$ for $CeNi_4Si$. It is noticed that this model does not describe well the low-temperature part of $S(T)$. The deviation between the experimental and theoretical data is presumably due to the presence of the antiferromagnetic correlations, which usually emerge as a minimum in $S(T)$ curve. As shown in Fig. 1b, the above-mentioned model gives a satisfactory fit to the data of the $YbNi_4Si$ compound. For $YbNi_4Si$, the fit parameters are $\Delta = -0.11$ meV and $\Gamma = 10$ meV, with $c = -0.0345$. The

negative value for $YbNi_4Si$ indicates that DOS peak is supposedly just below the Fermi level. A tail of the $4f_{7/2}$ peak of the Yb^{2+} spin–orbit split doublet is visible in XPS [1].

4. Summary

We have measured and analysed the temperature dependence of TEP for $CeNi_4Si$ and $YbNi_4Si$ compounds. This dependence for Ce and Yb compounds with the valence fluctuation is explained by the two-band conductor model assuming a single Lorentzian $4f$ band. The width of the narrow band Γ is of the same order of magnitude as that of the Ce and Yb valence fluctuation compounds. Nevertheless, a smaller value of Γ in the case of $YbNi_4Si$ may be responsible for a larger TEP in comparison with $CeNi_4Si$.

We have found that the position of the DOS in the investigated Ce and Yb-based compounds lies in opposite side in respect of the Fermi level.

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