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Thermoelectric Power in Ce Systems with Unstable Valence

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Abstract: In this paper, we report on a few exemplary tests of the applicability of analysis based on the interconfiguration fluctuation model (ICF) for a description of the temperature dependence of the thermoelectric power, $S(T)$. The examples include a series of alloys: $CeNi_2(Si_{1-y}Ge_y)_2$, $Ce(Ni_{1-x}Cu_x)_2Si_2$, and the fluctuating valence (FV) compound $CeNi_4Ga$. The two series develop from $CeNi_2Si_2$ being the FV system, where the f states occupation increases progressively with the Ge or Cu substitution. We find here that the ICF model parameters are of similar magnitude both for the analysis of the temperature dependence of the magnetic susceptibility and thermoelectric power. The ICF-type model appears to be a powerful tool for the analysis of $S(T)$ dependences in Ce-based FV compounds and alloys.

Keywords: thermoelectric power; fluctuating valence; Ce compounds

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1. Introduction

In recent years, widespread research has been directed towards waste heat recovery, i.e., making use of the heat dissipated in the environment. One of the ways is to make use of the thermoelectric effect. A thermoelectric device has important advantages over other classical heating/cooling solutions. It has no moving parts, can be integrated with typical electronic circuits, it provides a possibility of precise local cooling and heating, and finally, it is environmentally friendly. The main challenge is to improve the efficiency of thermoelectric devices expressed by the figure of merit $ZT = S^2\sigma T/\kappa$, where S is the Seebeck coefficient, σ denotes the electrical conductivity, and κ is the thermal conductivity. ZT can be increased, for example, by: reducing the lattice thermal conductivity [1], increasing the charge carrier effective mass (magnetic doping) in systems such as Bi_2Te_3 [2] or enhancing spin fluctuations [3]. The Seebeck effect in f-electron systems has been intensively studied for decades due to the high density of electronic states at the Fermi level, especially for intermetallics based on lanthanides such as cerium. It stems from the relationship between the thermoelectric power (TEP) and the density of states, D, namely $S \propto T(\partial lnD/\partial E)_{E_F}$.

However, it is not only the potential applications that drive researchers' efforts, but also the need to better understand the mechanisms governing the temperature dependence $S(T)$. The proposed theories range from advanced approaches such as those based on the Coqblin-Schrieffer model [4,5] to two-band models [6–9]. Recently, Stockert et al. [10] proposed a direct relation of thermoelectric power to temperature changes of valence $v(T)$, without going into the microscopic mechanisms. The thermoelectric power has been expressed by the approximate equation:

$$S = S_{ref} + S_{VF} = aT - b\frac{dv}{dT} \tag{1}$$

where the first term corresponds roughly to a reference material with a stable valence. Additionally, the overall formula describes the thermoelectric power of a single channel. We treat a and b as free parameters.

Moreover, as stems from the above discussion, an estimation of $S(T)$ should be possible if the temperature dependence of valence $v(T)$ is known. Furthermore, it is suspected to be the main source of the changes in the number of accessible charge carriers. The occupation of the f states, $1 - v(T)$, can be obtained from spectroscopic studies, but most often it is extracted from the magnetic susceptibility $\chi(T)$ analyzed in the framework of the interconfiguration fluctuation (ICF) model [11,12]. This model assumes that the configurations $4f^{\mathrm{n}}$ and $4f^{\mathrm{n}-1}$ differ in energy by E_{ex} and the rate of fluctuations ω_{SF} is related to the spin fluctuation temperature $T_{\mathrm{SF}} = \hbar\omega_{\mathrm{SF}}/k_{\mathrm{B}}$. In the formulation of the ICF susceptibility, $\chi_{\mathrm{ICF}}(T)$, an effective temperature is used either in the form $T_{\mathrm{eff}} = T + T_{\mathrm{SF}}$ [11,12] or $T_{\mathrm{eff}} = (T^2 + T_{\mathrm{SF}}{}^2)^{1/2}$ [13–15]. Therefore, one finally gets:

$$\chi_{\mathrm{ICF}}(T) = \frac{N_{\mathrm{A}}\mu_{\mathrm{eff}}^2[1 - v(T)]}{3k_{\mathrm{B}}T_{\mathrm{eff}}} \tag{2}$$

where N_{A} is the Avogadro's number, the effective paramagnetic moment for cerium is $\mu_{\mathrm{eff}} = 2.54\ \mu_{\mathrm{B}}$, k_{B} is the Boltzmann constant, and the mean occupation of the ground (nonmagnetic) state, $\nu(T)$, is expressed by:

$$\nu(T) = \frac{1}{1 + 6exp[-E_{\mathrm{ex}}/T_{\mathrm{eff}}]} \tag{3}$$

with E_{ex} in units of temperature. Finally, Equation (1) can be rewritten as:

$$S = S_{ref} + S_{VF} = aT - b\frac{-6TE_{\mathrm{ex}}exp[-E_{\mathrm{ex}}/T_{\mathrm{eff}}]}{(1 + 6exp[-E_{\mathrm{ex}}/T_{\mathrm{eff}}])^2 T_{\mathrm{eff}}^{3/2}} \tag{4}$$

for the case of $T_{\mathrm{eff}} = (T^2 + T_{\mathrm{SF}}{}^2)^{1/2}$, and

$$S = S_{ref} + S_{VF} = aT - b\frac{-6E_{\mathrm{ex}}exp[-E_{\mathrm{ex}}/T_{\mathrm{eff}}]}{(1 + 6exp[-E_{\mathrm{ex}}/T_{\mathrm{eff}}])^2 T_{\mathrm{eff}}^{2}} \tag{5}$$

for the case of $T_{\mathrm{eff}} = T + T_{\mathrm{SF}}$.

The model of Stockert et al. [10] for the description of the temperature dependence of the thermoelectric power, $S(T)$, has been proposed and verified for the example of Eu-based compounds, namely $\mathrm{EuNi_2P_2}$ and $\mathrm{EuIr_2Si_2}$. They related $S(T)$ to the temperature dependent Eu valence via Equation (1). One can expect that it should work for many other systems with fluctuating valence (FV). In the present paper, we use a similar approach for the exemplary Ce-based compounds characterized by the well-established FV state. However, in each case, we assume that the temperature dependent Ce valence is well-represented by the ICF model. In the Results section, the applicability of Equation (5) will be tested for some Ce-based compounds, for which we have previously observed the presence of the fluctuating valence. The aim of the present study is to verify if the ICF-type model of the thermoelectric power provides reliable values of fit parameters and, in respect to magnetic susceptibility, it can be applied as an alternative characteristic of the FV state. Furthermore, the effectiveness of the current model can make it complementary to the two-band models [6–9].

2. Materials and Methods

The discussed exemplary compositions were studied and described in our previous papers [16–21], where the details concerning the synthesis of the polycrystalline samples by arc-melting and the measurements of the thermoelectric power can be found. The main point of the current study is to show how the novel model of $S(T)$ works for these exemplary compounds. To carry out the new analysis of the previous data, we implemented Formulas (4) and (5) into the program OriginPro 8 SR0, v8.0725, (OriginLab Corporation, Northampton, MA, USA).

3. Results

In the following subsections, Equation (5) will be used for the analysis of the temperature dependence of the thermoelectric power, which we measured previously for some Ce-based compounds. The choice of the tested compounds and alloys stems from our previous extended studies, which confirmed the fluctuating valence state for these compositions.

3.1. $CeNi_2(Si_{1-y}Ge_y)_2$

$CeNi_2Si_2$ is a fluctuating valence (FV) system [6,16,22], whereas $CeNi_2Ge_2$ is a heavy fermion (HF) compound with the Kondo temperature of about 30 K [16,23–26]. We have recently investigated the physical properties for compositions in the transition range between the terminal compounds, i.e., for the alloys $CeNi_2(Si_{1-y}Ge_y)_2$. We have shown that for a small amount of Ge, especially $y \leq 0.25$, the fluctuating valence implies large values of the Kondo temperature. Additionally, the temperature dependence of TEP can be well described by the single-ion Kondo model [16]. However, the analysis of the magnetic susceptibility using the ICF model has shown that the change of valence with temperature is still meaningful even for y up to 0.63 [16]. Therefore, the $CeNi_2(Si_{1-y}Ge_y)_2$ series is a good candidate to test the range of applicability of the Stockert model. Figure 1 shows magnetic susceptibility for y equal to 0.13 and 0.63 and fitted using the ICF model as we have already presented in Ref. [16]. Moreover, it is shown that a similar behavior can be expected for y = 0.25 and 0.75. The obtained values of the parameters are listed in Table 1 in brackets, which include the results obtained with the assumption of $T_{eff} = (T^2 + T_{SF}{}^2)^{1/2}$. Figure 2 presents a refinement of the thermoelectric power dependence on temperature, $S(T)$, using Equation (5). It is evident that similar to the case of the magnetic susceptibility, a good fit is obtained for y up to 0.63 and the effectiveness of the model starts to drop for y = 0.88. From the results in Table 1, the values of T_{SF} decrease with the increasing Ge content, which very well reflects the expected damping of the valence fluctuations.

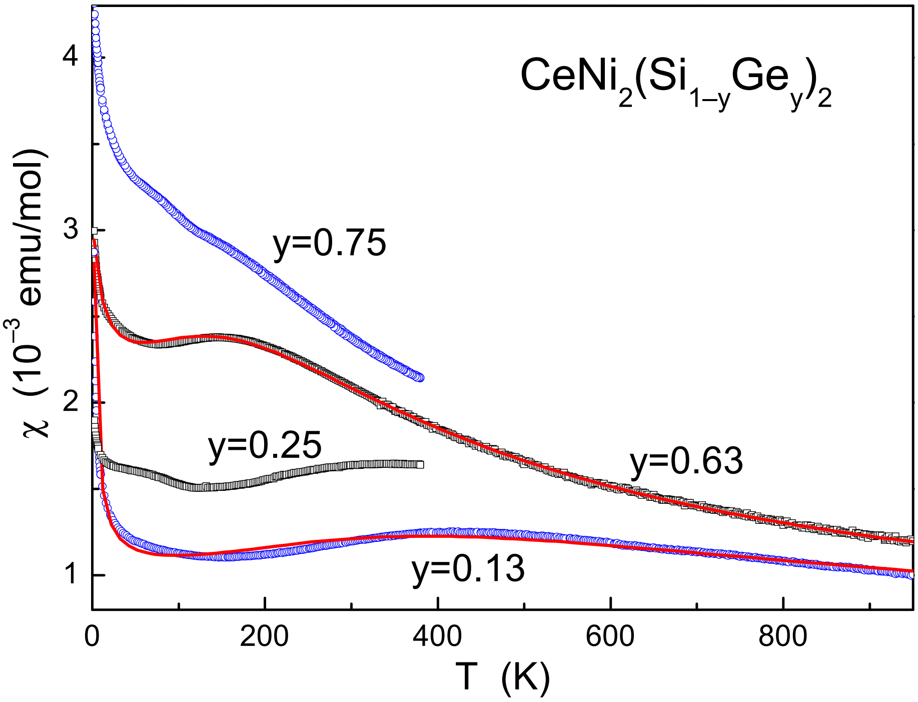

Figure 1. Magnetic susceptibility of $CeNi_2(Si_{1-y}Ge_y)_2$. The ICF model is applied for y = 0.13 and 0.63 (measured up to 1000 K), Equation (2).

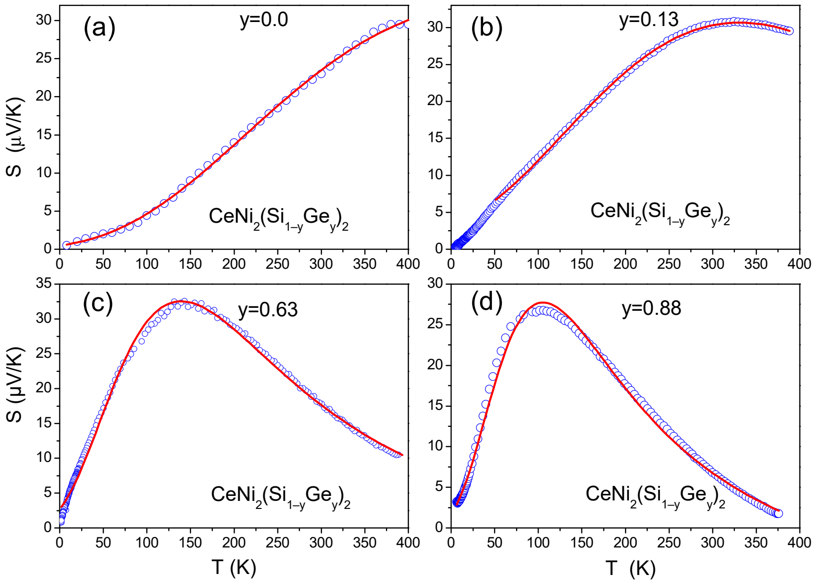

Figure 2. Thermoelectric power as a function of temperature fitted with Equation (5). (**a**) y = 0.0, (**b**) y = 0.13, (**c**) y = 0.63, (**d**) y = 0.88. The obtained values of the parameters are gathered in Table 1. $S(T)$ measurements were described in Ref. [16].

Table 1. Spin fluctuation temperature T_{SF} and interconfigurational excitation energy E_{ex} for thermoelectric power of CeNi$_2$(Si$_{1-y}$Ge$_y$)$_2$ analyzed by Equation (5) (with $T_{eff} = T + T_{SF}$).

y (Ge)	T_{SF} (K)	E_{ex} (K)
0.0	242	2455
0.13	259 (265 ^{a}, 310 ^{b})	2094 (1196 ^{a}, 960 ^{b})
0.63	96 (209 ^{a}, 154 ^{b})	768 (465 ^{a}, 399 ^{b})
0.88	68	569

^{a} TEP parameters derived from Equation (4) assuming $T_{eff} = (T^2 + T_{SF}{}^2)^{1/2}$. ^{b} Parameters derived from the analysis of the magnetic susceptibility by the ICF model assuming $T_{eff} = (T^2 + T_{SF}{}^2)^{1/2}$, data from [16].

To plot the occupancy of the *f* states as a function of temperature, we assume following Refs. [27–30], that the highest possible contribution of the Ce 4f^0 state is 0.30. Therefore, (1 − 0.3$v(T)$) is presented in Figure 3. For comparison, the *f* occupancy, which we have previously obtained at RT from the analysis of the Ce 3*d* XPS spectrum [16] with the procedure proposed by Gunnarsson-Schönhammer-Fuggle [31,32], is about 0.8 for y = 0.

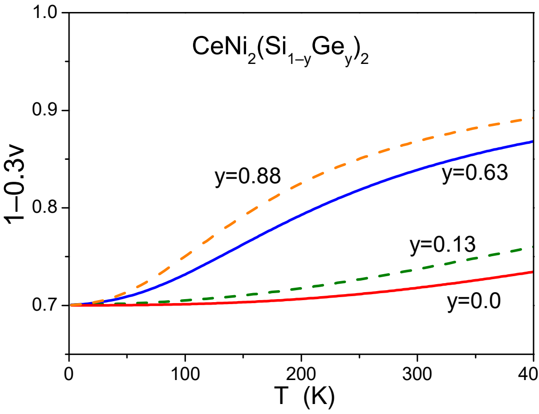

Figure 3. Occupancy of the *f* states as a function of temperature derived from the thermoelectric power $S(T)$ for CeNi$_2$(Si$_{1-y}$Ge$_y$)$_2$.

3.2. $Ce(Ni_{1-x}Cu_x)_2Si_2$

The next example concerns the $Ce(Ni_{1-x}Cu_x)_2Si_2$ series of alloys. This time, the valence state is modified by the substitution on the site of the 3*d* element. For example, the fluctuation of valence in $CeNi_2Si_2$ is progressively suppressed by the Cu substitution for Ni, where the terminal $CeCu_2Si_2$ compound is a heavy fermion system with T_K ~ 10 K [17]. The crystal structure is tetragonal of the $ThCr_2Si_2$-type for the entire range of x. Figure 4 shows the temperature dependence of the thermoelectric power fitted with Equation (5) for the Cu content x equal to 0.25 and 0.88. The spin fluctuation temperature T_{SF} and the interconfigurational excitation energy E_{ex} take the values 168 and 1450 K for the former and 82 and 769 K for the latter, respectively. However, our previous analysis of the magnetic susceptibility with the ICF model [17] for x = 0.25 was carried out for the case of $T_{eff} = (T^2 + T_{SF}{}^2)^{1/2}$ and provided T_{SF} = 205 K and E_{ex} = 766 K. It corresponds to Equation (4) for the TEP, which provides comparable values of T_{SF} = 258 K and E_{ex} = 1097 K. Figure 5 illustrates the temperature dependences of the *f* occupancy determined by Equation (5).

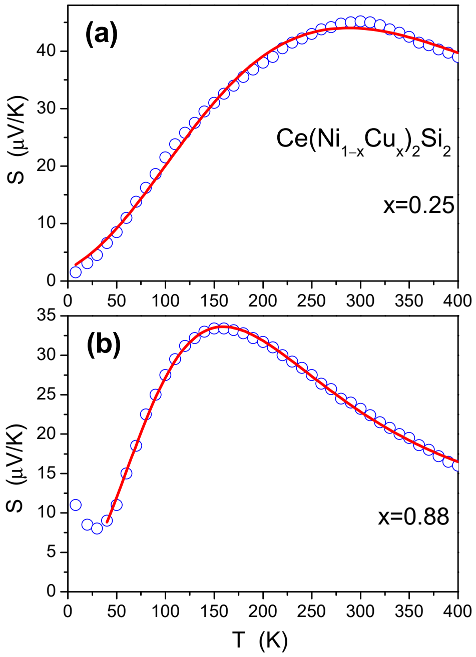

Figure 4. Thermoelectric power of $Ce(Ni_{1-x}Cu_x)_2Si_2$ as a function of temperature fitted with Equation (5). (**a**) x = 0.25 and (**b**) x = 0.88. See the text for the values of the parameters.

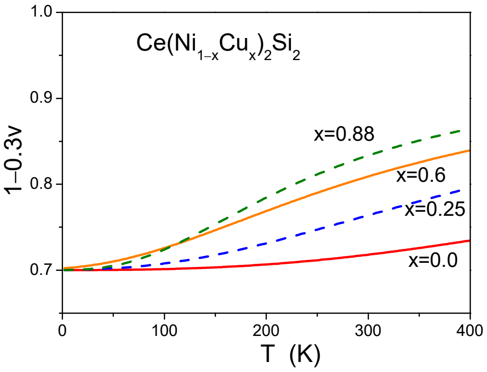

Figure 5. Occupancy of the *f* states as a function of temperature derived from the thermoelectric power $S(T)$ for $Ce(Ni_{1-x}Cu_x)_2Si_2$.

3.3. $CeNi_4Ga$

$CeNi_4Ga$ crystallizes in the hexagonal $CaCu_5$-type structure, with space group $P6/mmm$. We classified this compound as being in the fluctuating valence (FV) state [18–20], with the occupancy of the f states $n_f = 0.76$ and the hybridization of the f states with the conduction electrons $\Delta = 68$ meV. The temperature dependence of TEP for $CeNi_4Ga$ is negative in the whole temperature range studied [18], which results from the partly unfilled Ni 3d states and the reduced contribution of the Ce f states at the Fermi level. These characteristics of $CeNi_4Ga$ make it the next suitable candidate for testing the effectiveness of the TEP model based on the temperature dependence of the valence. The analysis with Equation (5) provides the red solid line shown in Figure 6 and the obtained values of the parameters are $T_{SF} = 97$ K and $E_{ex} = 1203$ K. Again, it is interesting to compare the resulting occupancy of the f state with estimates by other methods. On the right-hand axis of Figure 6, the temperature dependence of the f occupancy is plotted in a similar manner as for the previous exemplary compounds. The value at 300 K is equal to 0.75, which is in perfect agreement with the value $n_f = 0.76$, which we have obtained by the analysis of the RT Ce 3d XPS spectrum [21].

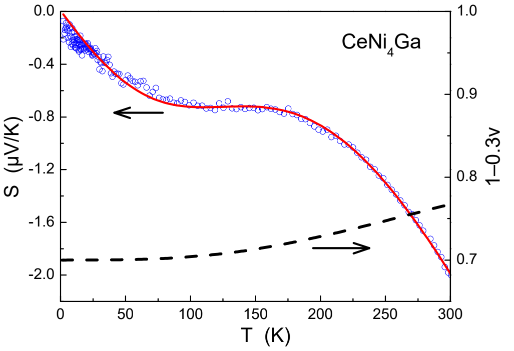

Figure 6. Left axis: Thermoelectric power $S(T)$ of $CeNi_4Ga$ fitted with Equation (5). See the text for the values of the parameters. Right axis: Occupancy of the f states as a function of temperature.

4. Discussion and Summary

Most of the models of the thermoelectric power in intermetallics, based on anomalous lanthanides such as Ce or Yb, refer to magnitudes such as density of states at the Fermi level, electronic band structure, and other quantities, which, in fact, include indirectly the inherent property of these systems—the non-integer valence state. Therefore, the application of the model, making direct use of the temperature dependence of the f states occupancy, seems fully justified. The presented examples corroborate the effectiveness of the approximation proposed by Stockert et al. [10]. We have shown that the ICF parameters, previously derived from the temperature dependence of magnetic susceptibility, are comparable with the results obtained in the present paper from the temperature dependence of thermoelectric power. Nevertheless, some differences in the parameters values are obviously apparent, thus a question arises: Which experimental method is more reliable? It should be remembered that the magnetic susceptibility is very sensitive to even a small amount of magnetic impurities, which are often difficult for quantitative extraction. Therefore, the data obtained from the analysis of $S(T)$ may be more resistant to such effects.

In summary, we gain a unique opportunity to realize a combined analysis of the magnetic susceptibility and thermoelectric power for fluctuating valence systems. Enhancements of the Seebeck parameter, possibly related to the unstable valence have been experimentally reported also for many compounds containing Yb (i.e., [33,34]). Therefore, the discussed model can also be effective for Yb-based systems. Furthermore, the valence state may provide an important contribution to the thermoelectric power in some Sm-based compounds [35].

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