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Determination of the crystal-field parameters for CePtSn

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Abstract. We have determined a set of crystal-field parameters for CePtSn which accounts for the available susceptibility, high-field magnetization at 4.2 K and inelastic neutron-scattering data. The analysis of experimental data revealed that the single-ion approach is reliable mainly at high temperatures ($T \geq T_N, T_K$) or at high magnetic fields where the energy associated with the exchange and coherence effects is much lower.

1. Introduction

Among the cerium intermetallic compounds, the equiatomic ternary systems CeTX ($T \equiv$ transition metal; $X \equiv$ p metal) are currently receiving wide scientific interest and they are the subject of systematic investigations [1]. In particular, CePtSn was classified as a ‘metallic Kondo compound’, which orders antiferromagnetically at 7.5 K [2]. A previous investigation on a single crystal of CePtSn has revealed strong anisotropy of the magnetic and transport properties and an attempt was made to construct the magnetic phase diagram [3]. The anisotropy of the magnetic susceptibility was successfully described by suitable chosen parameters in the microscopic crystal-field (CF) Hamiltonian, but the deduced CF level splitting of the $^2F_{5/2}$ multiplet ($\Delta_1 = 304$ K; $\Delta_2 = 573$ K) does not agree with the results of recent inelastic neutron-scattering (INS) experiments, which provide values of $\Delta_1 = 273$ K, $\Delta_2 = 425$ K [4] or $\Delta_1 = 278$ K, $\Delta_2 = 405$ K [5].

On the other hand, the precise determination of CF parameters is highly desirable in order to enable a quantitative interpretation of transport properties and the low-temperature properties in general. The single-ion approach of CF theory is bound to break down at low temperatures, where the coherence and magnetic ordering effects prevail. The periodic Anderson model (see, e.g., [6, 7]), involving the lowest CF doublet, is likely to give the best description in this regime. The nature of this doublet involved in this description is determined by the CF Hamiltonian, which, in turn, can be established by analysing some of the physical properties in high magnetic fields and at high temperatures, where the single-ion approach is adequate. In this paper, we therefore present a full analysis of published magnetic data, specific-heat and INS measurements in order to determine a consistent set of CF parameters.

2. Method of analysis

The high-temperature ($T > T_N; T_K \simeq 10$ K) properties of CePtSn are governed by a strong CF interaction which splits the 4f-electron states of the Ce^{3+} ion [3]. The environment

of the Ce site has approximately C_s point symmetry if the approximate space group $Pnma$ is used instead of the correct orthorhombic space group $Pn2_1a$ [8]. To keep the number of the adjustable parameters in the CF Hamiltonian to a minimum, we assume orthorhombic symmetry of the CF interaction used by Takabatake *et al* [3]. The microscopic CF Hamiltonian of the required orthorhombic symmetry is usually written in the Stevens parametrization scheme [9];

$$\mathcal{H}_{CF} = B_2^0 O_2^0 + B_2^2 O_2^2 + B_4^0 O_4^0 + B_4^2 O_4^2 + B_4^4 O_4^4 \quad (1)$$

where B_L^M are the CF parameters, which describe the strength of the interaction, and O_L^M are the operator equivalents. In the following, we shall determine the values of the CF parameters by an analysis of experimental data. For this purpose, it is convenient to introduce the Walter parametrization scheme [10]:

$$\mathcal{H}_{CF} = W[(1 - |x_1| - |x_2| - |x_3| - |x_4|)O_2^0/3 + x_1 O_2^2/2 + x_2 O_4^0/60 + x_3 O_4^2/12 + x_4 O_4^4/12]. \quad (2)$$

The parameters x_1 , x_2 , x_3 and x_4 are confined to the interval $(-1, +1)$ and are chosen in such a way that $|x_1| + |x_2| + |x_3| + |x_4| \leq 1$. The parameter W is a scaling factor, which can be easily deduced from the known full splitting $\Delta_2 = 425$ K of the ${}^2F_{5/2}$ manifold. The usual procedure to use this parametrization scheme is to prepare the computer code which allows one to move through the (x_1, x_2, x_3, x_4) parameter space with small increments δ_i in each parameter ($\delta_i = 0.01$ is usually sufficient), to calculate the desired physical quantities at such a well defined mesh and to compare with the experimental results available [11]. In comparison with any available fitting procedure, this method is numerically stable and allows in general unambiguous determination of the CF parameters.

A CF of orthorhombic symmetry splits the ${}^2F_{5/2}$ ground-state multiplet into three Kramers doublets of Γ_5 symmetry [12]. The estimated magnetic entropy up to the 40 K is close to $R \ln 2$, confirming that the double degenerate CF ground state is well isolated on the energy scale. Furthermore, the entropy value close to $R \ln 2$ indicates a Kondo temperature T_K close to or smaller than T_N . There are two possible magnetic-dipole transitions starting from this doublet and both of them were unambiguously identified by INS spectroscopy [4, 5].

Figure 1 displays the magnetization curves taken at 4.2 K in fields along the three principal axes [3]. For $B \parallel a$, the curve shows a change in slope at around 9 T, which can be ascribed to a metamagnetic transition connected with the destruction of the antiferromagnetic ground state. After the metamagnetic transition, the magnetization increases further and attains a value of about $1.2\mu_B$ /formula unit in the highest field of 35 T. The magnetizations measured for $B \parallel b$ and $B \parallel c$ show an almost linear increase with increasing field. Since the splitting of the CF levels at 35 T is large compared with the characteristic energy of intersite magnetic interaction estimated from the ordering temperature $T_N = 7.5$ K, we may expect to be able to reproduce the magnetization values M_a , M_b and M_c in terms of a single-ion CF scheme. Therefore, we use these values of magnetization as input to the parameter-search program.

The temperature dependence of the magnetic susceptibility measured along the three principal axes shows considerable anisotropy, which becomes more pronounced with decreasing temperature, indicating a significant influence of the CF interaction [3]. The high-temperature slopes of χ_i^{-1} against T provide values of the paramagnetic Curie temperatures for each crystallographic direction: $\theta_a = -35$ K, $\theta_b = -25$ K and $\theta_c = -145$ K. On the basis of molecular-field theory [13], the values of θ_i can be related to the values of

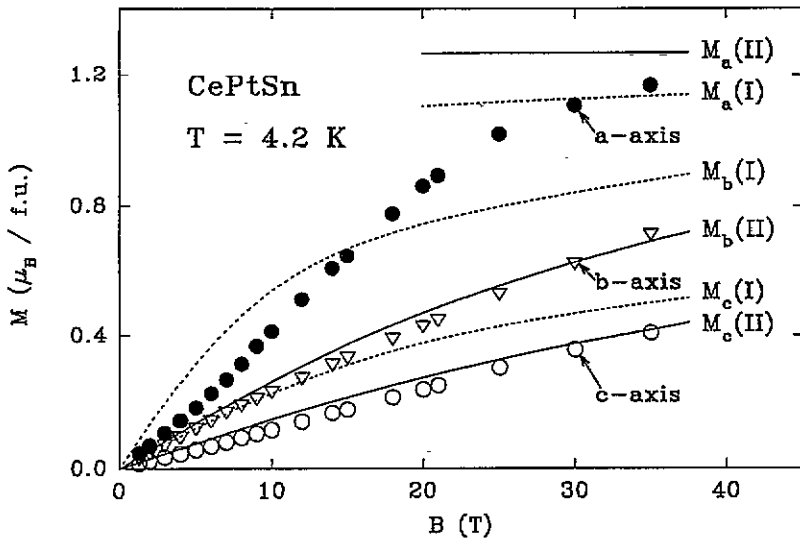


Figure 1. Experimental (\bullet , ∇ , \circ) and calculated (---, CF parameter set I [3]; —, CF parameter set II) field dependences of magnetization of CePtSn at 4.2 K, along the three principal directions. For the a axis, only the high-field parts of the calculated magnetization curves are displayed.

the CF parameters $B_2^0 \simeq 11$ K and $B_2^2 \simeq 1.5$ K. We choose the crystallographic a and c axes as the x and z quantization axes, respectively. Inspecting equations (1) and (2) we find that both values of W and x_1 should be positive. Furthermore, the crossing of the $\chi_a(T)$ and $\chi_b(T)$ dependences near 150 K, which may serve as a further test for the CF parameters, should be noted. Takabatake *et al* [3] analysed $\chi(T)$ using the expression $\chi^{-1}(T) = \chi_{\text{CF}}^{-1}(T) - \lambda$, where χ_{CF}^{-1} is the single-ion-based CF contribution and λ describes the effective intersite magnetic interaction. Using this, these authors derived satisfactory agreement with $\lambda = -199 \times 10^3$ mol m $^{-3}$ and the CF parameters denoted as set I in table 1. However, this set I provides values of M_a , M_b and M_c at 35 T (table 2) which are not in agreement with experiment.

Using the CF Hamiltonian (2) and the parameter-search method, we were able to determine values of W , x_1 , x_2 , x_3 and x_4 which account for the energies and the approximate relative intensities of the two observed INS transitions, the magnetization values M_a , M_b and M_c at 35 T as well as the anisotropy of magnetic susceptibility at suitable chosen temperatures ($T = 20, 80, 140$ and 200 K in our particular case). Our numerical search proved that there is only one area of the reduced parameter space satisfying the experimental results summarized in table 2 ($W > 0$, $x_1 > 0$, $x_2 > 0$, $x_3 < 0$ and $x_4 > 0$). Having identified this area, we repeated the search in this area with the step $\delta_i = 0.005$. The best solution is represented by $W = 159$ K, $x_1 = 0.02$, $x_2 = 0.17$, $x_3 = -0.23$ and $x_4 = 0.44$ corresponding to CF parameters denoted as set II in table 1. The area of possible solutions is rather small ($0.0 < x_1 < 0.04$, $0.15 < x_2 < 0.18$, $-0.26 < x_3 < -0.22$ and $0.41 < x_4 < 0.45$), mainly because of the requirement to reproduce the magnetization data at 35 T.

Having determined the CF parameters, we compare the measured M - B curves for the b and c directions with the calculated curves. The calculation of the magnetization curves has been done by means of the single-ion Hamiltonian

$$\mathcal{H} = \mathcal{H}_{\text{CF}} - g_J \mu_B \mathbf{B} \cdot \mathbf{J} \quad (3)$$

Table 1. CF parameters of CePtSn deduced from susceptibility only (set I; taken from [3]) and deduced from INS, susceptibility and high-field magnetization (set II). Also the corresponding wavefunctions are given.

CF parameter	Value for the following sets	
	Set I	Set II
B_2^0	8.5	7.4
B_2^2	0.24	1.6
B_4^0	-0.15	0.45
B_4^2	-4.1	-3.05
B_4^4	8.7	5.83
ψ_0	$+0.57 -\frac{1}{2}\rangle - 0.60 +\frac{3}{2}\rangle + 0.56 -\frac{5}{2}\rangle$ $+0.57 +\frac{1}{2}\rangle - 0.60 -\frac{3}{2}\rangle + 0.56 +\frac{5}{2}\rangle$	$-0.42 +\frac{1}{2}\rangle + 0.78 -\frac{3}{2}\rangle - 0.47 +\frac{5}{2}\rangle$ $+0.42 -\frac{1}{2}\rangle - 0.78 +\frac{3}{2}\rangle + 0.47 -\frac{5}{2}\rangle$
ψ_1	$-0.82 -\frac{1}{2}\rangle - 0.49 +\frac{3}{2}\rangle + 0.31 -\frac{5}{2}\rangle$ $-0.82 +\frac{1}{2}\rangle - 0.49 -\frac{3}{2}\rangle + 0.31 +\frac{5}{2}\rangle$	$+0.88 -\frac{1}{2}\rangle + 0.47 +\frac{3}{2}\rangle - 0.02 -\frac{5}{2}\rangle$ $-0.88 +\frac{1}{2}\rangle - 0.47 -\frac{3}{2}\rangle + 0.02 +\frac{5}{2}\rangle$
ψ_2	$+0.09 -\frac{1}{2}\rangle - 0.63 +\frac{3}{2}\rangle - 0.77 -\frac{5}{2}\rangle$ $+0.09 +\frac{1}{2}\rangle - 0.63 -\frac{3}{2}\rangle - 0.77 +\frac{5}{2}\rangle$	$-0.21 -\frac{1}{2}\rangle + 0.42 +\frac{3}{2}\rangle + 0.88 -\frac{5}{2}\rangle$ $-0.21 +\frac{1}{2}\rangle + 0.42 -\frac{3}{2}\rangle + 0.88 +\frac{5}{2}\rangle$

Table 2. Experimental and calculated energy-level splitting and magnetization values at 35 T and 4.2 K for the three principal directions using the CF parameter set I (taken from [3]) and set II (see table 1). In parentheses we quote the magnetization values calculated including ferromagnetic exchange interaction in the molecular-field approximation ($\lambda = -199 \times 10^3 \text{ mol m}^{-3}$).

	Experiment	Set I	Set II
Δ_1 (K) [4]	273	304	264
Δ_2 (K) [4]	425	573	425
Δ_1 (K) [5]	278	—	—
Δ_2 (K) [5]	405	—	—
M_a (μ_B /formula unit)	1.17	1.13	1.26(1.27)
M_b (μ_B /formula unit)	0.71	0.87	0.69(0.71)
M_c (μ_B /formula unit)	0.41	0.50	0.41(0.43)

where g_J is the Landé factor and J the total angular momentum. The magnetic field B contains contributions from the applied field and the dipolar and exchange interactions of the Ce^{3+} ion with its neighbours in the mean-field approximation [14]. The latter contributions should be small in the present case but can be dominant in systems like RFe_2 for example [15]. The Hamiltonian (3) yields the following expression for the magnetization:

$$M = g_J \mu_B \sum_i \langle \eta_i | J | \eta_i \rangle \frac{\exp(-\epsilon_i / k_B T)}{Z} \quad (4)$$

where η_i and ϵ_i are the i th eigenvector and eigenvalue, respectively, of the Hamiltonian (3) and Z is the partition function. We found satisfactory agreement with the experimental data for the b and c directions (see figure 1), which is improved compared with the results obtained with set I of CF parameters. In our calculations this originates from the dominant weights of the $|\pm\frac{3}{2}\rangle$ states in the ground-state wavefunction ψ_0 (see table 1). The influence of the exchange and dipolar interaction on the calculated curves can be estimated by introducing the molecular-field parameter λ found by Takabatake *et al* [3]. It was found that the calculated values of magnetization at 35 T changed by less than 5% (see table 2). This result encouraged us to calculate the high-field magnetization curves in the paramagnetic range ($T = 18$ and 30 K) and we find a rather strong curvature for the a direction (figure 2).

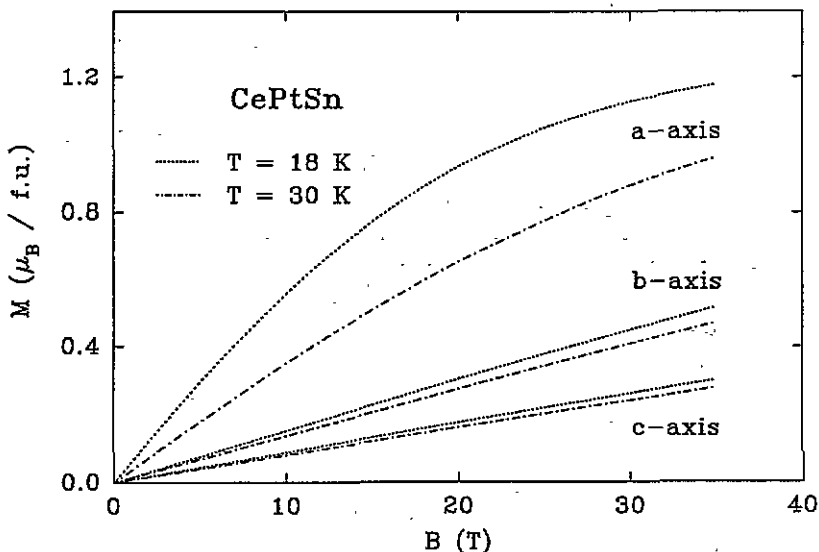


Figure 2. Calculated field dependence of the magnetization of CePtSn for the three principal directions at 18 K (.....) and 30 K (- · -) using CF-parameter set II.

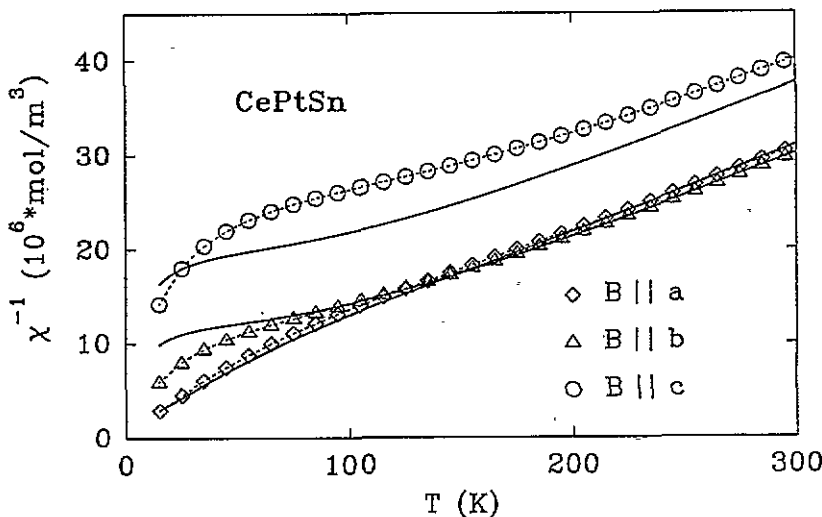


Figure 3. Calculated temperature dependence of the magnetic susceptibility of CePtSn along the three principal directions using the CF-parameter set I (- - -, taken from [3]), and set II (—). The symbols denote the particular crystallographic directions.

It would be of interest to measure the high-field magnetization in the paramagnetic range in order to test our CF parameter set II further.

Furthermore, we compared the calculated temperature dependence of the magnetic susceptibility of our set II with that obtained by set I [3] (figure 3). The characteristic features of the susceptibility of CePtSn ($\chi_a > \chi_b > \chi_c$ at low temperatures, and the crossing of $\chi_a(T)$ and $\chi_b(T)$ at about 150 K) are well reproduced also by set II, although absolute agreement with the experimental data is less good than for set I.

3. Discussion and conclusions

The main purpose of this work was to determine a consistent set of CF parameters for the intermetallic compound CePtSn. We have shown that a full analysis of the experimental data (INS, magnetization and susceptibility) leads to a CF Hamiltonian reproducing these data in a satisfactory way. In [3], only the susceptibility data are taken into account, but there are at least two arguments to suggest that more attention is paid to the high-field magnetization data. First, we may assume that, as in other CeTX compounds, the coherence effects as well as the exchange interaction mainly influence the low-field ($B < 20$ T) magnetization data. In this context we can understand why, for the a axis, only the high-field parts of the experimental and theoretical magnetization curves (as shown in figure 1) approach reasonable agreement. Secondly, the ground-state CF doublet of the Ce^{3+} ion in CePtSn is split by $\Delta_a = 59.4$ K, $\Delta_b = 14.8$ K and $\Delta_c = 10.3$ K upon application of a magnetic field of 35 T in the a , b and c directions, respectively. These splittings exceed T_N and $T_K \simeq 10$ K [3], which can be taken as measures of the exchange and coherence effects, respectively. Therefore the use of the single-ion Hamiltonian (3) can be justified for these high fields as well as for $T \gg T_N, T_K$. This is supported by a recent successful analysis of the more complex intermetallic compound CeCu₂Si₂ where it was shown that a simple CF model is adequate to describe most of the observed features at temperatures well above the Kondo temperature, which is about 10 K in this compound [16].

Acknowledgments

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