



Electronic and crystal structure of α - and β -CeIr₂Si₂

M. Mihalik*, M. Diviš, V. Sechovský

Charles University, Faculty of Mathematics and Physics, Department of Condensed Matter Physics, Ke Karlovu 5, 121 16 Prague 2, Czech Republic

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ABSTRACT

We present the results of XRD, magnetization, resistivity and specific heat measurements of CeIr₂Si₂ single crystals for both, the low-temperature α -phase and the high-temperature β -phase, respectively. The α -phase adopts the tetragonal ThCr₂Si₂-type whereas the β -phase forms in the CaBe₂Ge₂-type structure. Both the phases remain paramagnetic down to low temperatures, nevertheless both, the magnetization and resistivity exhibit pronounced anisotropy in the whole temperature range of measurements (2–300 K). Results of fitting the temperature dependence of the susceptibility within the interconfiguration-fluctuation model point to the Ce valence fluctuating between 3+ and 4+. The α -phase behaves as a Fermi-liquid (FL) at low temperatures whereas the β -phase exhibits non-Fermi-liquid (NFL) features. The results are discussed in context of other similar polymorphic compounds.

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Up to now, there are several known polymorphic systems, characterised by the high-temperature tetragonal CaBe₂Ge₂-type phase and the low-temperature tetragonal ThCr₂Si₂-type phase. The RENi₂As₂ (RE = La, Ce, Pr, Nd and Sm), REIr₂Si₂ (RE = La and Ce) compounds [1–3] and UCo₂Ge₂ [4] serve as examples. Niepmann and Pöttgen [3] report on CeIr₂Si₂ of the crystallographic structure for both the phases whereas Hiebl et al. [5] point to different magnetic behaviors of the two phases and speculate about the intermediate valence state for the β -phase. The lack of further information motivated us to prepare single crystals of both, the α - and β -CeIr₂Si₂, and to measure the details into magnetic, transport and thermal properties.

The CeIr₂Si₂ single crystal has been grown by the Czochralski method in a tri-arc furnace. As a starting material we have used a stoichiometric melt of the elements of at least 3 N purity and a tungsten rod as a seed. We have found that the crystals can be grown by a pulling speed as high as 14 mm/h and that there is no need for necking during the pulling procedure. The drawback of the pulling is that the grown ingots always brake after about 25 mm of length, which is presumably due to the onset of the β - α phase transformation: the volume of the two phases is identical, but the c/a ratio differs substantially [3,5]. The change of the c/a ratio probably induces large internal stresses and consequent cracks formation in the material causes the final rod rupture. Nevertheless, we were able to extract the part of the grown ingot, which we have found to be a single crystal of the β -phase. This has been confirmed by a Laue pattern and by an X-ray powder pattern. Part of the grown single-crystalline ingot was wrapped into the tantalum foil, sealed into the quartz tube under Ar protective

atmosphere and annealed at 900 °C for 12 h. The analysis of the annealed sample (X-ray powder diffraction, Laue pattern) confirmed the single crystal being the α -phase. We have also performed the EDX analysis of both the samples (of the α - and β -phase, respectively) which confirmed the proper 1:2:2 stoichiometry and absence of any spurious phase.

For the comparative analysis of the specific heat, we have also prepared a polycrystalline sample of LaIr₂Si₂ by arc melting (the β -phase). The α -phase of LaIr₂Si₂ has been obtained by the same annealing procedure as in case of CeIr₂Si₂. Both, the α - and β -LaIr₂Si₂ samples have been also checked by the X-ray powder diffraction and by the EDX.

The Rietveld analysis of the obtained X-ray powder patterns of CeIr₂Si₂ provided the crystallographic parameters $a = 0.4084(5)$ nm; $c = 1.015(2)$ and $c/a = 2.485$ for the α -phase and $a = 0.4144(4)$ nm; $c = 0.9850(1)$ nm and $c/a = 2.377$ for the β -phase, respectively, which are in very good agreement with previously published data [3,5].

The specific heat (C), resistivity (ρ) and magnetoresistivity were measured in the PPMS (Quantum Design) system. For the $C(T)$ measurements, we have used plate-shape samples of a typical mass ~ 15 mg. The resistivity and magnetoresistivity were measured on bar-shape samples of a typical cross section of 0.6 mm² and a length of ~ 3 mm. The magnetization measurements were performed using the SQUID magnetometer (MPMS system—also from Quantum Design). These experiments were done on bar-shaped samples with typical mass of around 30 mg. The magnetic field was always applied along the longest side of the bar. We have not corrected our data for the demagnetization factor, because of the very low magnetization of the sample.

Above 25 K, the $C(T)$ dependence for CeIr₂Si₂ fits very well to its La-based analogue for both, the α - and β -phase (Fig. 1). This implies that the phonon contribution to the specific heat is similar

* Corresponding author. Tel./fax: +420 2 2191 1352.

E-mail address: mihalik@mag.mff.cuni.cz (M. Mihalik).

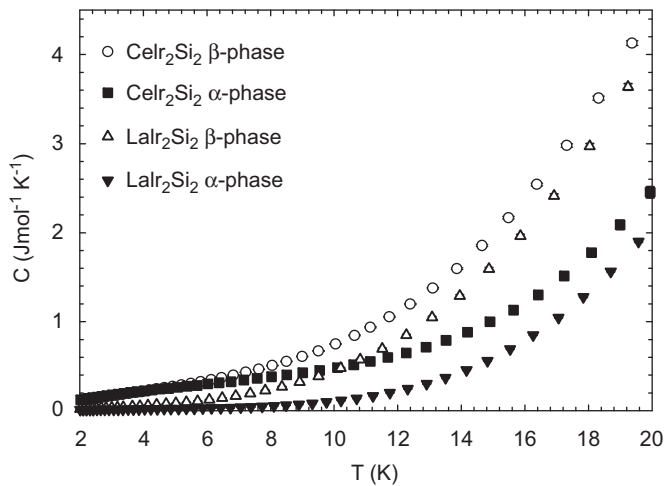


Fig. 1. The comparison of the low-temperature detail of both phases of CeLr_2Si_2 and LaLr_2Si_2 specific heat.

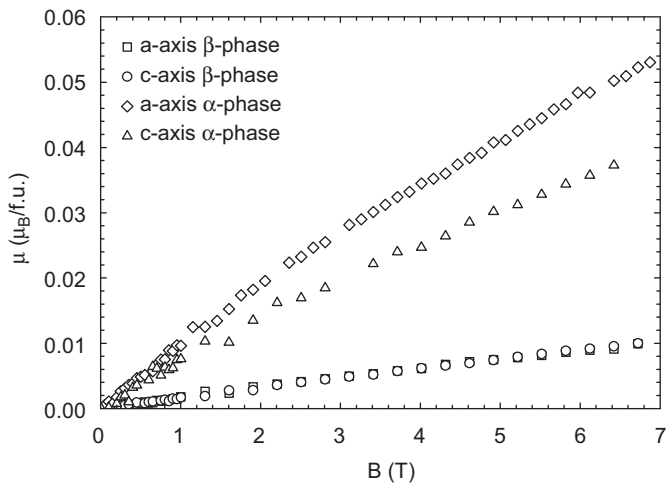


Fig. 2. The magnetization curves for both phases measured at 2 K.

in the both compounds. At lower temperatures the corresponding specific heat values for CeLr_2Si_2 are considerably higher than that for LaLr_2Si_2 . The $C(T)$ dependence measured for $\beta\text{-CeLr}_2\text{Si}_2$ is linear for $T < 6$ K. For the α -phase, we have observed enhancement of the C values with temperature decreasing below 5 K. The C/T ratio at $T = 2$ K reaches $60 \text{ mJ mol}^{-1} \text{ K}^{-2}$ at 2 K, which implies an enhanced value of the γ -coefficient.

The magnetization curves are plotted for both the phases in Fig. 2. For the α -phase the magnetization curves are slightly curved and their corresponding magnetization values are higher than for the β -phase. Nevertheless, all the magnetization curves are linear at magnetic fields higher than 3 T. For the magnetic susceptibility determination, we have measured the $M(T)$ curves in magnetic fields $B_1 = 3$ T and $B_2 = 5$ T and from these curves we have calculated the magnetic susceptibility as $(M_2 - M_1)/(B_2 - B_1)$. The inverse magnetic susceptibility (Fig. 3) exhibits similar maxima and minima as the susceptibility data for YbB_4 , $\text{U}_2\text{Ru}_2\text{Sn}$ and U_2RuGa_8 [6,7]. Since the studied compounds are considered to be the interconfiguration-fluctuation compounds and also Hiebl et al. [5] claim, that Ce in CeLr_2Si_2 might be in the intermediate valence state, we have assumed that the Ce valence fluctuates between 3+ and 4+. According to the interconfiguration-fluctuation model proposed by Sales and Wohlfleben [8] and later modified by Franz et al. [9], we have

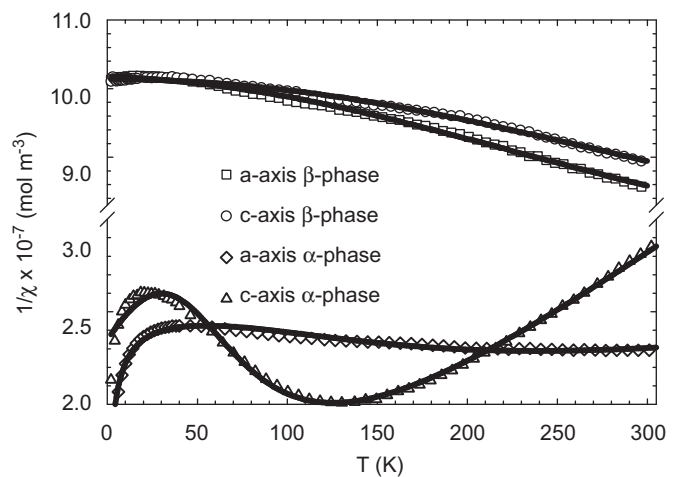


Fig. 3. The inverse susceptibility measured for the both phases and for field applied along both main crystallographic axes. The lines represent the best fit due to the interconfiguration fluctuation model.

Table 1

The summary of parameters obtained from the fitting of the magnetic susceptibility.

	n	E_{ex} (K)	T_f (K)	C ($\text{m}^3 \text{mol}^{-1}$)	θ_p (K)	χ_0 ($\text{m}^3 \text{mol}^{-1}$)
a-axis β -phase	0	-2190	250	-	-	9.8×10^{-9}
c-axis β -phase	0	-1810	380	-	-	8.6×10^{-9}
a-axis α -phase	2×10^{-5}	-590	192	2×10^{-3}	-2	-2.79×10^{-8}
c-axis α -phase	1×10^{-3}	-390	51	0.04	-154	-1.4×10^{-9}

successfully fitted the magnetic susceptibility by equation

$$\chi(T) = (1 - n) \left(\frac{\mu P(T)}{\sqrt{T^2 + T_f^2}} \right) + n \frac{C}{T - \theta_p} + \chi_0 \quad (1)$$

where

$$P(T) = \frac{2J + 1}{2J + 1 + \exp(-E_{ex}/k_B \sqrt{T^2 + T_f^2})} \quad (2)$$

and T_f is the spin-fluctuation temperature, n splits all the Ce ions to those, which are in the interconfiguration state (first term in equation) and in the normal Ce^{3+} state (second term in equation)—these ions are considered to obey Curie–Weiss law, respectively; The temperature-independent susceptibility χ_0 may be attributed, at least in part, to conduction electrons; $J (= 5/2)$ is the total angular momentum for the Ce^{3+} and E_{ex} is the excitation energy between the interconfiguration states. In Eq. (1) we have already assumed $J = 0$ for the Ce^{4+} ion. We were able to fit all measured susceptibility data with Eq. (1). The results of the fitting are summarized in Table 1. In the all four cases $E_{ex} < 0$, which implies the Ce^{3+} configuration lying higher in energy than the Ce^{4+} configuration. The coefficient n suggests that the majority of the Ce ions in the α -phase is in the interconfiguration-fluctuation state. For the β -phase all the Ce ions are in the interconfiguration-fluctuation state ($n = 0$).

Fig. 4 demonstrates the electrical resistivity of the β -phase. For this phase, we have found a broad bump of the c -axis $\rho(T)$ curve at temperatures around 200 K, which is a frequent phenomenon for the Ce-based intermetallic compounds [10,11]. The a -axis resistivity decreases monotonously with decreasing temperature and it is approximately 5 times lower than the c -axis resistivity. At low temperatures (< 15 K) the resistivity measured along both the main crystallographic axes of the β -phase varies linearly with

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