Journal of Magnetism and Magnetic Materials 322 (2010) 12–18

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Low temperature thermodynamical properties of $ErCu_2Si_2$

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ARTICLE INFO

Article history:
Received 19 May 2009
Available online 27 August 2009

PACS:
61.12.−q
75.25.+z
75.50.Ee

Keywords:
Magnetically ordered material
Magnetic susceptibility
Specific heat
Electrical resistivity
Neutron diffraction

ABSTRACT

$ErCu_2Si_2$ crystallises in the tetragonal $ThCr_2Si_2$-type crystal structure. In this paper results of magnetometric, electrical transport, specific heat as well as neutron diffraction are reported. Results of electrical resistivity and specific heat measurements performed at low temperature yield existence of magnetic ordering roughly at 1.3 K. These results are in concert with neutron diffraction measurements, which reveal simple antiferromagnetic ordering between 0.47 and 1.00 K. At temperatures ranging from 1.00 up to 1.50 K an additional incommensurate magnetic structure was observed. The propagation vector $\boldsymbol{k} = (0;0;0.074)$ was proposed to describe magnetic reflections within the amplitude modulated magnetic structure. Basing on specific heat studies the crystal field levels splitting scheme and magnetic entropy were calculated.

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1. Introduction

The $ErCu_2Si_2$ compound belongs to the group of materials with the tetragonal $ThCr_2Si_2$-type crystal structure [1]. The structural and magnetic properties of RCu_2Si_2 compounds, where R is a rare earth element, were published recently in a number of papers [2–13] including the determination of macroscopic magnetic properties [3,4], magnetic structure [5–8] and crystal field (CF) parameters [9–13].

Magnetometric measurements revealed that $ErCu_2Si_2$ is a paramagnet down to 4.2 K [3], while the authors of Ref. [4] predicted the antiferromagnetism occurring below 5 K. ^{166}Er Mössbauer measurements hinted at antiferromagnetic ordering below 1.65 K [12]. Simultaneously to our studies two papers, concerning specific heat and magnetic properties of $ErCu_2Si_2$ single crystals, have been published [14,15]. The specific heat data collected at low temperature range for $ErCu_2Si_2$ gave two anomalies at 1.50 and 0.98 K [14], being in excellent accordance with our results of neutron diffraction reported in this paper. The other paper brings magnetic susceptibility as well as resistivity studies [15].

In this paper the results of comprehensive investigation including set of complementary experimental techniques on polycrystalline $ErCu_2Si_2$ sample are reported. The obtained data are discussed in context of literature data both for $ErCu_2Si_2$ and whole RCu_2Si_2 family.

2. Experimental details

Polycrystalline sample of $ErCu_2Si_2$ was prepared by arc melting of the constituents (Er: 99.9 wt%; Cu: 99.99 wt%; Si: 99.99 wt%) under a titanium-gettered argon atmosphere. The melted ingot was subsequently annealed in an evacuated silica tube at 870 K for 100 h. The quality of the resulting product was checked by X-ray powder diffraction at the room temperature using the Philips PW-3710 X'PERT diffractometer (CuKα radiation).

The DC magnetometric measurements were carried out in the 1.7–400 K temperature range in magnetic fields up to 5 T using the Quantum Design MPMS-5 SQUID magnetometer. The electrical resistivity was measured over the 0.4–300 K temperature interval using standard four contact DC technique. The specific heat data was investigated within the 0.4–400 K temperature range by relaxation method using the Quantum Design PPMS platform.

The neutron diffraction patterns were recorded at E6 diffractometer located at the BENSC (Helmholtz–Zentrum Berlin). The

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doi:10.1016/j.jmmm.2009.08.024

incident neutron wavelength was 2.452 Å. The powdered sample was enclosed within the cylindrical copper container of the diameter of 8 mm. Small amount of deuterated methanol was added into the container in order to ensure appropriate thermal contact between the sample grains. The data were collected at several temperatures between 0.47 and 1.60 K using the standard cryostat with the ^{3}He insert in it. The refinements were done by means of the Rietveld method using the FULLPOF software (version 4.40 August 2008) [16].

3. Experimental results and discussion

3.1. Crystal structure

The X-ray diffraction data collected at the room temperature as well as the neutron diffraction data confirmed unambiguously the tetragonal $ThCr_2Si_2$-type structure (I4/mmm). In this structure the atoms occupy the following sites:

2 Er atoms at the 2(a) site (0,0,0), (1/2, 1/2, 1/2)
4 Cu atoms at the 4(d) site (1/2,0,1/4), (0,1/2,1/4), (1/2,0,3/4), (0,1/2,3,4)
4 Si atoms at the 4(e) site (0,0,*z*), (0,0,-*z*), (1/2,1/2,1/2+*z*), (1/2,1/2,1/2-*z*)

The values of the lattice parameters and the free positional parameter *z*, at room temperature (XRD) and 1.60 K (neutron diffraction) are listed in Table 1.

3.2. Magnetometric studies

In Fig. 1 results of magnetometric investigations of $ErCu_2Si_2$ are presented. The inverse magnetic susceptibility varies linearly with temperature above roughly 100 K. Fitting the Curie–Weiss law to these data yields the effective magnetic moment $\mu_{\mathrm{eff}} = 9.18(1)\,\mu_B$, i.e. it is slightly smaller than the theoretical free Er^{3+} ion value (9.58 μ_B). The paramagnetic Curie temperature is close to zero, which suggests that magnetic correlations between the Er magnetic moments are very weak. Below 100 K some departure from the straight line behaviour of $\chi^{-1}(T)$ is observed, presumably due to crystal field effect. The compound remains paramagnetic down to the lowest reached temperature of 1.72 K, as inferred from featureless susceptibility in the low-temperature region and characteristic behaviour of the isothermal magnetisation (see the insets of Fig. 1).

3.3. Electrical resistivity measurements

As displayed in Fig. 2, electrical resistivity of $ErCu_2Si_2$ behaves in a metallic manner with quasi-linear temperature dependence above 100 K. When lowering the temperature some saturation of the resistivity is observed below roughly 50 K, whereas below 10 K, a sizeable downturn in $\rho(T)$ occurs. The latter feature may be attributed to diminishing spin-disorder resistivity due to crystal field effect or/and short-range magnetic correlations. Though the temperature derivative of the resistivity does not exhibit any clear singularity in the low-temperature region (note the lower inset of Fig. 2), the local maximum at 1.3 K seems to be related to the magnetic ordering, as evidenced by specific heat measurements (as follows).

In the paramagnetic region, the electrical resistivity can be described by the Bloch–Grüneisen–Mott (BGM) formula (solid line in Fig. 2):

$$\rho(T) = (\rho_0 + \rho_\infty) + 4RT\left(\frac{T}{\theta_D}\right)^4 \int_0^{\theta_D/T} \frac{x^5 dx}{(e^x - 1)(1 - e^{-x})} - KT^3 \qquad (1)$$

where the first term stands for the scattering of conduction electrons on static defects in the crystal lattice (the residual

Table 1
Refined structural parameters of $ErCu_2Si_2$ together with reliability factors at the room temperature (XRD) and 1.60 K (neutron diffraction).

T (K)	300	1.60
a (Å)	3.9390(6)	3.932(1)
c (Å)	9.985(2)	9.994(5)
c/a	2.535(1)	2.542(2)
V (Å^3)	154.9(1)	154.5(2)
z	0.377(2)	0.379(2)
$R_{profile}$ (%)	8.9	1.7
R_{Bragg} (%)	9.5	5.6

Standard deviations are given in parentheses.

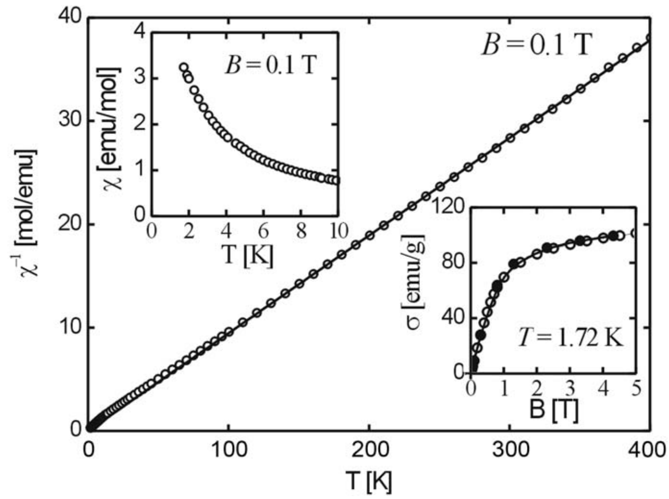

Fig. 1. Temperature dependence of the inverse magnetic susceptibility of $ErCu_2Si_2$. The solid line represents the Curie–Weiss fit discussed in the text. The upper-left corner inset displays the magnetic susceptibility at low temperatures. The bottom-right corner inset shows the isothermal magnetisation taken at 1.72 K with increasing (open symbols) and decreasing (filled symbols) magnetic field.

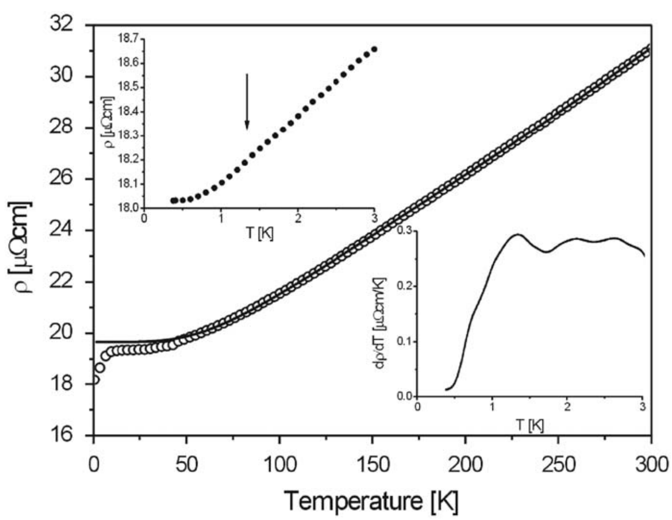

Fig. 2. Temperature variation of the electrical resistivity of $ErCu_2Si_2$. The solid line stands for the Bloch–Grüneisen–Mott fit discussed in the text. The insets show the low-temperature resistivity and its temperature derivative. The arrow aims the supposed magnetic transition.

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