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Magnetism and high-field magnetization of ErCu_2

K. Sugiyama^{a,b,*}, T. Yamamoto^a, N. Nakamura^a, A. Thamizhavel^a, S. Yoshii^b,
K. Kindo^b, N.H. Luong^{a,c}, Y. Ōnuki^{a,b}

^a Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan

^b Research Center for Materials Science at Extreme Conditions, Osaka University, Toyonaka, Osaka, 560-8531, Japan

^c Center for Materials Science, Viet Nam National University, Hanoi, 334 Nguyen Trai, Thanh Xuan, Hanoi, Viet Nam

Abstract

The magnetization of ErCu_2 has been measured in high magnetic fields up to 45 T. Metamagnetic transitions are found at 16.5, 0.7 and 13 T for the field applied along the a -, b - and c -axis, respectively. The sharp metamagnetic transition for $H\parallel b$ is due to the antiferromagnetic ordering, while the other two metamagnetic transitions originate from the crossover of the crystalline-electric-field energy levels of the 4f electrons.

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1. Introduction

The rare-earth intermetallic compounds RCu_2 have the orthorhombic CeCu_2 -type crystal structure, except LaCu_2 . These compounds have attracted a lot of interest because of their interesting magnetic properties [1,2]. One interesting property is the metamagnetic transition based on the quadrupolar interaction [3]. This phenomenon corresponds to the magnetic-anisotropy-axis conversion between the hard and easy axis in high magnetic fields, which can be called field-induced ferroquadrupolar ordering.

ErCu_2 is an antiferromagnet with Néel temperature $T_N = 11.3$ K and the b -axis as easy magnetic

axis [4]. Below T_N , transitions have been found at 6.1, 4.3 and 3.2 K in thermal-expansion, specific-heat and neutron-diffraction experiments [4], which are due to changes of the magnetic structure. The experimental results of Mössbauer spectroscopy, inelastic neutron scattering [5], the thermal-expansion [6] and the Schottky peak in the specific-heat [7] measurements have been analyzed on the basis of the crystalline electric field (CEF) scheme with the parameters set of $B_2^0 = -0.28$ K, $B_2^2 = -0.22$ K, $B_4^0 = -0.30 \times 10^{-2}$ K, $B_4^2 = -0.30 \times 10^{-2}$ K, $B_4^4 = -0.14 \times 10^{-2}$ K, $B_4^4 = -0.30 \times 10^{-2}$ K, $B_6^0 = -0.20 \times 10^{-4}$ K, $B_6^2 = -0.47 \times 10^{-4}$ K, $B_6^4 = -0.97 \times 10^{-4}$ K and $B_6^6 = -2.96 \times 10^{-4}$ K [5]. Previous measurements of the magnetization on a single crystal by Hashimoto et al. [1] were carried out in magnetic fields up to 5 T. As for the high-field magnetization, showing a metamagnetic transition with two steps, there is only one study on a polycrystalline sample in magnetic fields up to 25 T [2].

*Corresponding author. Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan. Fax: +81-6-6850-5372.

E-mail address: sugiyama@phys.sci.osaka-u.ac.jp (K. Sugiyama).

To clarify the metamagnetic transition in high fields, we have grown a single crystal and measured the high-field magnetization in a wide temperature range. The experimental results were analyzed on the basis of the CEF scheme.

2. Experimental

The single crystal was grown by the Czochralski pulling method in an induction furnace by using a tungsten crucible. The starting materials were 3N (99.9% pure)-Er and 5N-Cu. The single-crystal ingot was pulled at a speed of 10 mm/h under helium-gas atmosphere with 3.0 kg/cm^2 . The size of the ingot was 10 mm in length and 3 mm in diameter.

High-field magnetization measurements up to 45 T along the main three axes of the single crystal were carried out in the High Field Laboratory at Extreme Conditions, Osaka University, in pulsed fields with a pulse width of 20 ms. The magnetization was measured with a standard pick-up coil system. The magnetization in the steady fields up to 7 T and the magnetic susceptibility in the temperature range from 2 K to the room temperature were measured in a commercial SQUID magnetometer.

3. Experimental results and analysis

Fig. 1 shows the temperature dependence of the magnetic susceptibility. The *b*-axis corresponds to the magnetic easy axis and the sharp peak at 11.7 K for $H \parallel b$ is due to the occurrence of antiferromagnetic ordering below this temperature. The inset shows the inverse magnetic susceptibility, from which effective moments of 9.9 , 9.6 and $9.9 \mu_B$ are obtained for the field along the *a*-, *b*- and *c*-axis, respectively. These values are slightly large compared to the value of $9.58 \mu_B$ for the free Er^{3+} ion. The observed Weiss constants are -25 , 21 and -8 K for $H \parallel a$, *b* and *c*, respectively, and differ considerably from the values 18 , 53 and 36 K that have reported previously [1]. The solid lines in the inset indicate

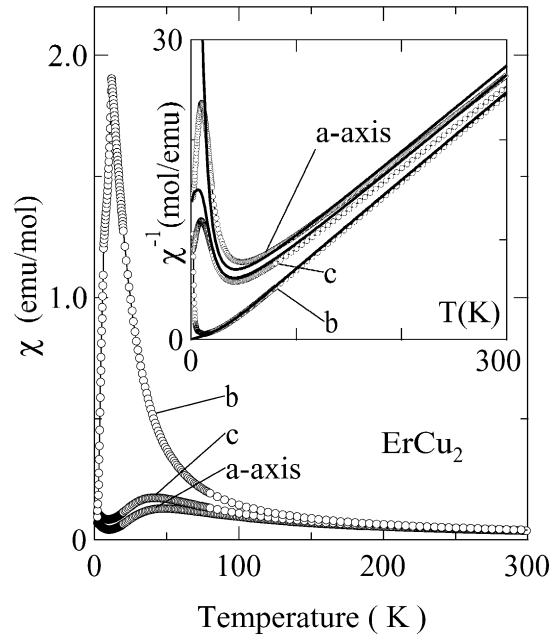


Fig. 1. Temperature dependence of the magnetic susceptibility of ErCu_2 . The inset shows the temperature dependence of the inverse susceptibility, where the solid lines correspond to the CEF curves.

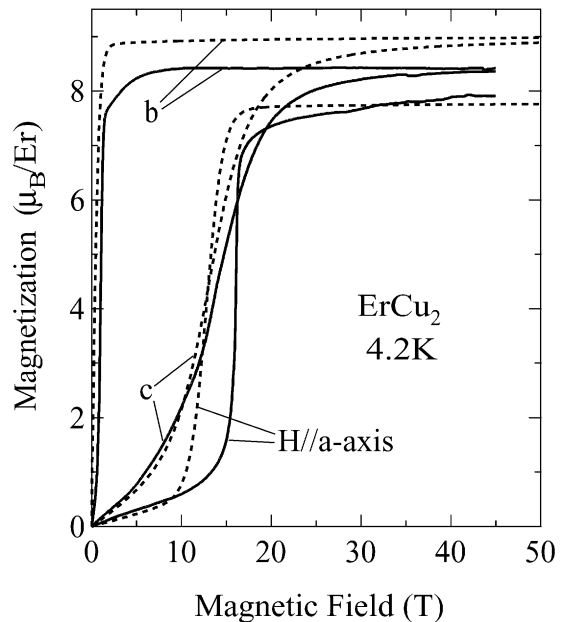


Fig. 2. High-field magnetization of ErCu_2 . The dotted lines correspond to the CEF curves.

the calculated CEF curves, obtained by using the CEF parameters mentioned in the Introduction [5]. The anisotropic susceptibility is well explained by this CEF scheme.

The magnetization at 4.2 K is highly anisotropic in low fields, as shown in Fig. 2. The magnetization for $H\parallel b$ (magnetic easy axis) shows a metamagnetic transition around 1 T and saturates at higher fields, similar to the previous results [1]. The saturation moment of about $8.4\mu_B/\text{Er}$ is close to the theoretical Er^{3+} free-ion value of $9\mu_B$. This metamagnetic transition is due to a field-induced change from the antiferromagnetic state into the forced-ferromagnetic state at high fields.

On the other hand, the metamagnetic transitions at 16 and 13 T for $H\parallel a$ and c (magnetic hard-axes), respectively, is not due to the antiferromagnetic

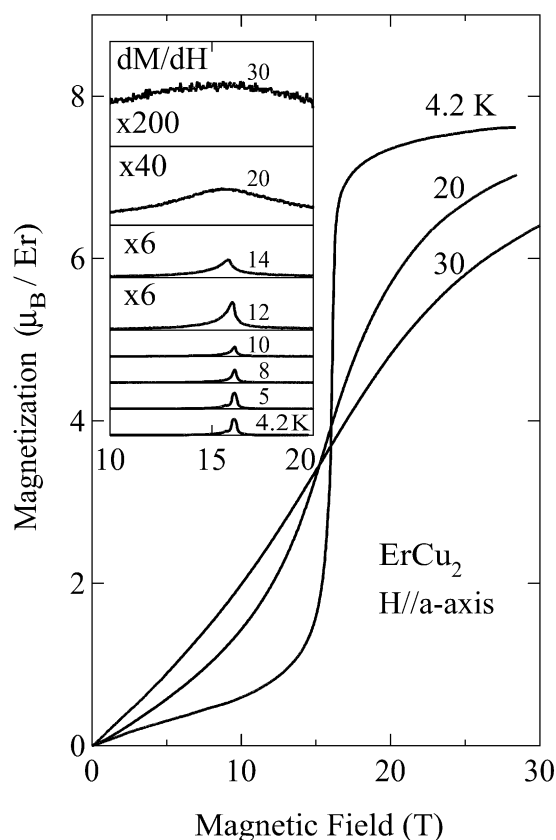


Fig. 3. High-field magnetization of ErCu_2 at various temperatures for $H\parallel a$. The inset shows the differential magnetization curves.

ordering, but due to crossing of CEF-levels. The three dotted lines in Fig. 2 correspond to the CEF magnetization curves that were calculated by using the same CEF parameters as mentioned above. The metamagnetic transitions for $H\parallel a$ and c are well explained by the present CEF scheme. We note that the average magnetization obtained along the three main axes is almost same as the previously reported polycrystalline magnetization [2].

To clarify experimentally the metamagnetic transition for $H\parallel a$, we have measured the magnetization for $H\parallel a$ in a wide temperature range, as shown in Fig. 3. The inset shows the differential magnetization dM/dH . A sharp peak is observed up to 14 K, which is close to the Néel temperature $T_N = 11.7\text{ K}$. Above 20 K, a broad metamagnetic behavior is still observed. Defining the transition field as the maximum of dM/dH , which corresponds to the maximum slope of the magnetization, the phase diagram is obtained that is shown in Fig. 4. The open circles correspond to the sharp metamagnetic transitions, while the open squares correspond to the broad metamagnetic transitions. At T_N , the sharp metamagnetic transition has changed into a broad transition and the transition field shifts with a small step of about 1 T to a slightly lower field value. This implies that the

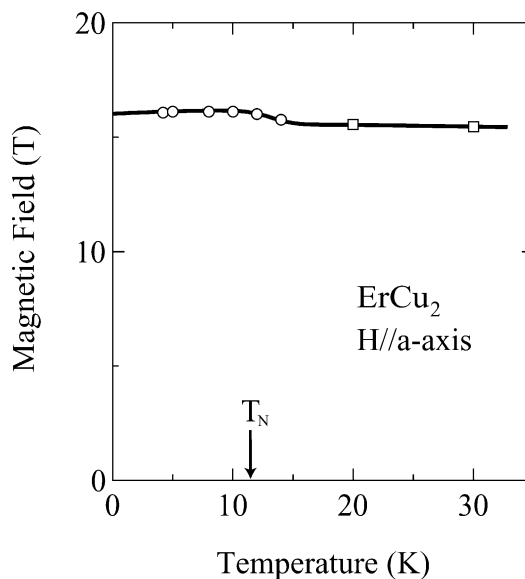


Fig. 4. Phase diagram of ErCu_2 for $H\parallel a$.

antiferromagnetic ordering effect on the CEF magnetization curve is not so large that the present CEF parameters would not be applicable at 4.2 K. It can thus be concluded that the present metamagnetic transition for $H\parallel a$ is not due to the antiferromagnetic ordering and can be well explained in terms of a level-crossing effect in the CEF scheme. Similar results are obtained for $H\parallel c$.

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References

- [1] Y. Hashimoto, H. Fujii, H. Fujiwara, T. Okamoto, J. Phys. Soc. Japan 47 (1979) 67, 73.
- [2] N.H. Luong, J.J.M. Franse, in: K.H.J. Buschow (ed.), Handbook of Magnetic Materials, Vol. 8, Elsevier Science Pub, Amsterdam, 1995. p. 415.
- [3] K. Sugiyama, M. Nakashima, Y. Yoshida, R. Settai, T. Takeuchi, K. Kindo, Y. Ônuki, Physica B 259–261 (1999) 896.
- [4] Y. Hashimoto, H. Kawano, H. Yoshizawa, S. Kawano, T. Shigeoka, Physica B 213 & 214 (1995) 333.
- [5] P.C.M. Gubbens, K.H.J. Buschow, M. Diviš, J. Lange, M. Loewenhaupt, J. Magn. Magn. Mater. 98 (1991) 141.
- [6] E. Gratz, M. Rotter, A. Lindbaum, H. Müller, E. Bauer, H. Kirchmayr, J. Phys.: Condens. Matter 5 (1993) 567.
- [7] N.H. Luong, J.J.M. Franse, T.D. Hien, N.P. Thuy, J. Magn. Magn. Mater. 140–144 (1995) 1133.