Field-induced magnetic ordering in ErNi\textsubscript{1-x}Cu\textsubscript{x}Al

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Abstract. The RNi\textsubscript{1-x}Cu\textsubscript{x}Al pseudoternaries belong to group of RTX intermetallic compounds crystallizing in the hexagonal ZrNiAl type of structure. In the critical region of the substitution parameter \( x \sim 0.8 \) there was found a loss of a long-range magnetic order at the lowest studied temperatures for series with \( R = \text{Tb, Dy, Er} \). As an explanation for this effect a competition of different ordering mechanisms for either RNiAl or RCuAl compounds was proposed. Nevertheless, the magnetization curves of the ErNi\textsubscript{0.2}Cu\textsubscript{0.8}Al compound (within the critical concentration region) showed behaviour close to the parent ErCuAl, which was confirmed as a ferromagnet. Neutron diffraction on ErNi\textsubscript{0.2}Cu\textsubscript{0.8}Al compound with applied magnetic field signifies appearing of the ferromagnetic order at the lowest applied magnetic field of 0.1 T. The magnitude of the ordered magnetic moment increases with increasing magnetic field in agreement with the magnetization curve, confirming tendency to ferromagnetic ordering or short-range magnetic order of ferromagnetic character in this critical region.

1. Introduction

The RNI\textsubscript{1-x}Cu\textsubscript{x}Al compounds belong to a large group of 1:1:1 \( RTX \) compounds (where \( R \) stands for a rare-earth element, \( T \) for a transition metal and \( X \) for a \( p \)-metal element) crystallizing in the hexagonal ZrNiAl-type of structure and exhibiting a large variety of magnetic structures in the ordered state. Very surprising results have been observed for the TbNi\textsubscript{1-x}Cu\textsubscript{x}Al series [1]. A surprising behaviour of the ordered magnetic moments on the position of rare-earth element \( R \) was found around specific concentration of the substituted Cu instead of Ni. Within past years we have performed structural and magnetic investigation for the ErNi\textsubscript{1-x}Cu\textsubscript{x}Al and DyNi\textsubscript{1-x}Cu\textsubscript{x}Al compounds including neutron powder diffraction. In this work we discuss our recent neutron-diffraction results on the ErNi\textsubscript{1-x}Cu\textsubscript{x}Al series under external magnetic field in the context with above-mentioned work and previously studied Er- and Dy-based series [2,3].

In all studied systems the loss of long-range magnetic order (LRO) is observed at the same Cu-concentration range between \( x = 0.6 \) to 0.8. That means that the rare-earth element species does not play considerable role in this effect. It is thus only the number of 3d-electrons in (Ni,Cu) sublattice that is the decisive parameter. It may be conceived with an idea of competition between different mechanisms responsible for magnetic order in RNI\textsubscript{Al} or RCu\textsubscript{Al}, respectively: RKKY interaction (Rudeman-Kittel-Kasuya-Yosida interaction referring to a coupling mechanism between localized...
moments through polarization of the conduction electrons) and exchange interaction mediated by transition-metal free \( d \)-electron states [1].

We have performed a neutron diffraction experiment on \( \text{ErNi}_{0.2}\text{Cu}_{0.8}\text{Al} \) compound with applied magnetic field in order to investigate magnetic structures induced by the magnetic field, and to clarify the similarity of the magnetization curves of the compound with lost LRO against the parent magnetically ordered one.

2. Experimental

The polycrystalline sample was prepared by arc-melting in a monoarc furnace under protection of argon atmosphere. The sample was then checked by X-ray diffraction for its quality. The sample was additionally checked for its phase composition by using a microprobe analysis using the JXA-733 electron microprobe microanalyzer. Pure Er, Ni, Cu and Al metals were used as standards. The weight concentrations were obtained from the measured relative intensities (i.e. ratios to the standards) using the ZAF method [4] and the SAMx program support [5]. Besides the main phase with \( 1:0.2:0.8:1 \) (Er:Ni:Cu:Al) stoichiometry (which corresponds to the nominal composition within accuracy of the used method) there were found also traces of pure Er and a small amount of foreign phase corresponding to composition \( \text{Er}_{0.2}\text{([Ni,Cu]})_{0.5}\text{Al}_{0.3} \). No reflection corresponding to the pure erbium was observable in the X-ray pattern, but two small excess peaks are visible on the X-ray pattern that most probably correspond to the last mentioned foreign phase. We shall mention that also boundary compound \( \text{ErCuAl} \) contained similar amount of the same impurities that have no considerable influence on the observed magnetic properties. As it was seen later by neutron diffraction (correspondingly also at present investigation), there was no foreign peak visible on the diffraction pattern that does not correspond to the \( \text{ZrNiAl}- \)type hexagonal structure, as well as not to other propagation than the ferromagnetic \((000)\) one, so we believe that the impurities do not influence magnetic behavior of our compound.

The neutron-diffraction experiment under magnetic field was performed at the Institute-Laue-Langevin Grenoble (ILL), using the D1B diffractometer working with a banana-type multidetector covering \( 80^\circ \). There was no collimation used only horizontal and vertical slits have been used before the cryostat in order to adjust the beam to the sample size. The a monochromatic beam of 2.52 Å has been selected from a thermal neutron beam by a highly oriented pyrolytic graphite monochromator (002 reflection), then to remove the contamination by higher harmonics a graphite filter has been used. The external magnetic field was used in the range up to 1.2 T.

The obtained diffraction data were analyzed by a whole-profile fitting - Rietveld refinement method, using the FullProf program [6] with corrections on instrumental abbreviations. The Fermi lengths and absorption coefficients were taken from Ref. [7].

3. Results and discussion

We have studied a sample with composition around the critical concentration of Cu (\( x = 0.8 \)) under applied magnetic field to find out changes induced by the applied magnetic field. The diffraction patterns taken at zero magnetic field in the paramagnetic state and at the lowest temperature (\( T = 15 \) and \( 1.5 \) K) are in good agreement with the previous results [2]. Under applied magnetic field new magnetic intensities on the top of the nuclear reflections appear already with very small magnetic field of \( 0.1 \) T, as it can be seen on Fig. 1. The induced magnetic moment at field of \( 0.1 \) T is \( 4.3 \, \mu_B \) (magnetic agreement factor \( R_{\text{Bragg}}^{\text{mag}} = 9.8 \)).
After application of the magnetic field of 0.35 T, the magnetic moment is 4.8 $\mu_B$ and under the magnetic field of 1.2 T the compound exhibits already the value of the ordered (ferro) magnetic moment of 6.8(4) $\mu_B$ ($R_B^{\text{mag}} = 7.2$) what corresponds to the observed magnetic moment in the parent ErCuAl [8]. The diffraction pattern (displayed on Fig. 1) clearly shows onset of magnetic contribution on position of the nuclear reflections, especially the (100) and (001) reflections increase very rapidly from almost zero value intensity to quite intense peaks.

Figure 2. The field dependence of the intensities of selected reflections of the ErNi$_{0.2}$Cu$_{0.8}$Al sample. Data are displayed for comparison together with magnetization curve in the same field range at similar temperature; also magnetization data of ErCuAl (taken from [8]) are displayed for comparison.

The evolution of intensities for several reflections with applied magnetic field at the lowest temperature of 1.5 K is displayed on the Fig. 2. A rapid onset of magnetic contribution to intensities of...
the reflections is visible on practically all of the reflections, representing onset of long-range magnetic ordering even at very small applied external field. The evolution of the integrated intensities (left part of Fig. 2) is corresponding to the field evolution of magnetization (right part of Fig. 2), where for comparison also magnetization data of ErCuAl are added. Note that ErCuAl was confirmed by neutron diffraction as a simple ferromagnet [8].

Previous assumption of magnetic order on the short range scale is most probably confirmed as well as its ferromagnetic character although its character at zero magnetic fields cannot be by principle estimated by this method of applying even very small magnetic field. Short range magnetic order caused probably by competition of two different ordering mechanisms in the concentration region around $x = 0.8$ (as proposed by Ehlers [1]) can be very easily removed by applying quite small magnetic field (0.1 T) what leads to ferromagnetic order of the magnetic moments ordered up to this point locally. This finding explains the inconsistency between the bulk magnetic data (magnetization curves indicating ferromagnetic behaviour of the compound [9]) and results from the neutron diffraction experiment performed on these compounds previously at zero external field which did not discover any magnetic contribution down to the lowest used temperature, except for enhanced intensities at low angles [10]. Confirmation of the ferromagnetic-type ordering on the local scale without applied magnetic field is going to be confirmed by the μSR experiment in near future.

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References

[1] Ehlers G, Ahlert D, Ritter C, Miekeley W and Maletta H 1997 Europhys. Let. 37(4) 269
[2] Prchal J, Javorský P, Sechovský V, Dopita M, Isnard O and Jurek K 2004 J. Magn. Magn. Mater. 283 34
[3] Prchal J, Javorský P, Detlefs B, Daniš S and Isnard O 2007 J. Magn. Magn. Mater. 310 e589
[4] Hulinský V and Jurek K Zkoumání látek elektronovým svazkem 1983 SNTL Prague
[5] KEVEX manual, SAMx manual
[6] Rodriguez-Carvajal J 1993 Physica B 192 55
[7] Sears VF 1992 Neutron News 3 26
[8] Javorský P, Burlet P, Ressouche E, Sechovský V, Michor H and Lapertot G 1996 Physica B 225 230
[9] Prchal J, Javorský P, Vejpravová J, Dopita M, Rafaj D, Sechovský V, Jurek K, Šantavá E and Maryško M 2003 Acta Physica Polonica B 34 1485
[10] Prchal J, Javorský P, Isnard O and Sechovský V 2004 Physica B 350 e159