Journal of Magnetism and Magnetic Materials 324 (2012) 3313–3322

Contents lists available at SciVerse ScienceDirect

Journal of Magnetism and Magnetic Materials

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

Magnetic disordered in Ti doped $ErCo_2$ alloys: Griffiths-like behavior

Y. Öner [a,*], M. Guillot [b]

[a] Department of Physics, Faculty of Science and Letters, Istanbul Technical University, 34469 Maslak, Istanbul, Turkey
[b] Laboratoire National des Champs Magnétiques Intenses, Grenoble Toulouse, CNRS, B.P. 166, 38042 Grenoble Cedex 9, France

ARTICLE INFO

Article history:
Received 27 December 2011
Received in revised form
12 May 2012
Available online 5 June 2012

Keywords:
Ferrimagnetic material
Intermetallic alloy
Magnetic phase transition
High magnetic field
Magnetic susceptibility
Electrical resistivity

ABSTRACT

We report electrical and magnetic properties of $ErCo_{(2-x)}Ti_x$ ($x=0$, 0.01, 0.02, 0.03, 0.04, 0.05) in the temperature range 4–300 K. The substitution of Ti for Co causes no change in the crystal structure of $ErCo_2$ (cubic Laves phase C15 with space group $Fd\bar{3}m$). The lattice parameter, a, decreases almost linearly with increasing x up to $x=0.04$ and then increases slightly for the largest Ti concentration. The Curie temperature, T_c is closely correlated with the lattice parameter. All resistivity curves exhibit a jump-like drop at T_c, which is the characteristic sign of the first order transition. The temperature dependence of the resistivity was analyzed using a conventional model for RCo_2 alloys. We have also investigated the role of the chemical disorder in the resistivity. The temperature dependence of the resistivity and the residual resistivity vs. x are interpreted consistently, considering the existence of some Co-rich ordered regions due to the quenched-in chemical disorder. Low-field magnetization measurements were carried out in the temperature range 4–100 K. The inverse susceptibility, $1/\chi$, shows non-linear behavior vs. temperature above T_c for all samples except $ErCo_2$. We used three different approaches to determine the magnetic moment per formula unit: modified Curie–Weiss type paramagnetism, Néel type paramagnetism, and Griffiths-like behavior. A detailed analysis of the magnetization at low-fields reveals the complexity of the magnetic state both in the ordered and paramagnetic phases. However, the findings in the paramagnetic regime suggest that an interpretation based on the formation of a Griffiths phase is most plausible. High-field magnetization (up to 23 T) was also investigated in a wide temperature range below and above T_c. In the paramagnetic regime, the magnetization data were described satisfactorily with the Landau theory. Landau coefficients a_1 and a_3 were determined as a function of temperature for selected samples. It was found that the coefficient a_3 shows a unique behavior for all the samples, which may be attributed to a characteristic of the mixed phase (dilute magnetic ordered domains in the paramagnetic regime).

1. Introduction

Within the RT_2 (R=Rare Earth, T=Transition Metal) families, the cubic Laves-phase RCo_2 compounds are of particular interest, since these compounds display interesting magnetic properties which arise from the coexistence and interaction of the well localized rare-earth 4f electrons and of the itinerant 3d electrons of Co. These compounds have been used as model systems for the magnetism of itinerant electron systems and itinerant electron metamagnetism (IEM) for decades. The first order transition is due to the itinerant electron metamagnetism occurring in the Co sublattice. The exchange field of ordered 4f moments in these compounds causes an abrupt increase in the Co magnetic moment from 0 to about 0.7−1.0 μ_B via the first order phase transition. (See the recent review by Gratz and Markosyan [1].) It is now well known that the inherent instability of Co sublattice magnetism is responsible for the occurrence of IEM. The electronic density-of-states (DOS) of the Co 3d-sublattice has a narrow peak in vicinity of the Fermi energy, which can be controlled by external factors such as temperature, pressure and impurities. The interrelation of the 3d-Co band structure and the IEM mechanism has prompted many researchers to carry out detailed magnetic and other related investigations in a variety of these compounds by using pressure or composition variations [2–4]. Although many of the experimental findings have been interpreted within the framework of itinerant magnetism, there is no definite consensus about how profoundly the electronic DOS features in vicinity of the Fermi energy and the external parameters are interrelated.

In recent years, it has been realized that Co magnetic clusters exist in the paramagnetic phase of $ErCo_2$ for a range of temperatures above the critical temperature T_c [5,6]. A recent work by Liu and Altounian [7] predicted theoretically a transition from a low-spin state for Co (0.1 μ_B) to the well known high-spin state of the

* Corresponding author. Tel.: +90 212 285 3260; fax: +90 212 285 6386.
E-mail address: oner@itu.edu.tr (Y. Öner).

0304-8853/$ - see front matter © 2012 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.jmmm.2012.05.044

phase (1 μ_B) at the onset of the ordering temperature of $ErCo_2$. The existence and nature of the Co moment in the paramagnetic phase of $ErCo_2$ unveiled a mostly ignored aspect of these systems. Recently, Herrero-Albillos et al. have reported that, using different experimental techniques such as X-ray magnetic circular dichroism (XMCD), small-angle neutron scattering (SANS), and ac magnetic susceptibility measurements [6,8,9], some dilute Co-rich regions exhibit a new magnetic order (referred to as a perimagnetic phase). This occurs on a nano-scale distance range due to antiferromagnetic interactions between Co moments and Er moments, which on average result in a net magnetic moments along the Co moment direction. Very recently, Herrero-Albillos et al. [10] have carried out detailed ac susceptibility measurements in order to elucidate this possible new phase. These studies above T_c in $ErCo_2$ point to the presence of Co disorder, for which the structure has not been identified unambiguously so far, nor has its relevance for the electronic and magnetic properties been studied. In view of these open issues, we have reinvestigated the magnetic properties of $ErCo_2$ and of the series of new Ti doped $ErCo_2$ compounds.

The main part of this study deals with the search for a Griffiths phase above T_c in $ErCo_2$ because of strong ferromagnetic coupling between dilute Co-rich regions in a paramagnetic matrix which might give rise to a new collective magnetic ordering between these dilute strong coupled Co-rich regions in the paramagnetic regime.

Since Griffiths singularities arise as a result of spatial fluctuation in the distribution of rare strongly coupled clusters in the disordered (paramagnetic) phase with quenched randomness, a study similar to that of $ErCo_2$ can be done on Ti doped $ErCo_2$, thereby demonstrating the coexistence in the paramagnetic phase of different sizes of magnetic clusters. In particular, the correlation length (varying exponentially with the cluster volume) can be determined vs. Ti concentration. Thus, this study may provide some of the most convincing evidence of a Griffiths-like phase in these materials. For this reason, we choose these materials and give evidence in favor of the existence of such a phase: Ti doped $ErCo_2$ compounds can be one of most compelling examples because the substituting Ti atoms are non-magnetic and have the same outer electronic shell configurations as those of the Co atoms. Ti atoms do not alter the crystal structure of $ErCo_2$ and their size is very close to that of Co atoms. In this work, we have systemically investigated the magnetic and electrical properties of $ErCo_{2-x}Ti_x$ for $x=0$, 0.01, 0.02, 0.03, 0.04 and 0.05 in a wide range of temperature (4–300 K) and magnetic field (in both low and high fields up to 23 T). Indeed, we have obtained some of the most convincing evidence yet for the interplay between, and the coexistence of, pure paramagnetism and dilute Co-rich ferromagnetic regions strongly coupled with each other above the critical temperature over a wide temperature range. Based on the magnetic and electrical measurements, we observed that Ti substitution introduces strong disorder as well. However, we also discuss alternative models to account for the temperature dependence of the magnetic susceptibility.

2. Experimental

The samples used in this study ($ErCo_{2-x}Ti_x$, $x=0.00$, 0.01, 0.02, 0.03, 0.04 and 0.05) were prepared from 99.9% pure starting materials by a conventional arc-melting technique. More detail concerning the preparation has been reported in our previous works [5]. Magnetization measurements at low fields were carried out using a commercial Physical Property Measurement System (PPMS, Quantum Design Model 6500) including a vibrating sample magnetometer (VSM) attachment. On the other hand, high magnetic field magnetization measurements were performed on the samples at the LNCMI (CNRS) laboratory in Grenoble under 23 T in a temperature range of 4.2–300 K using a sample extraction technique. We used a needle-shaped polycrystalline sample at low fields and its powder form at high-field studies. Resistivity of the samples was measured with a standard DC four-probe setup over a 5–300 K temperature range. Below 80 K, a calibrated Ge resistance thermometer (GR-200A-2500) allowed the measurement of the temperature while for higher temperatures a calibrated Pt thermometer was used. Electrical contacts were made using silver paint and 25 μm gold wire.

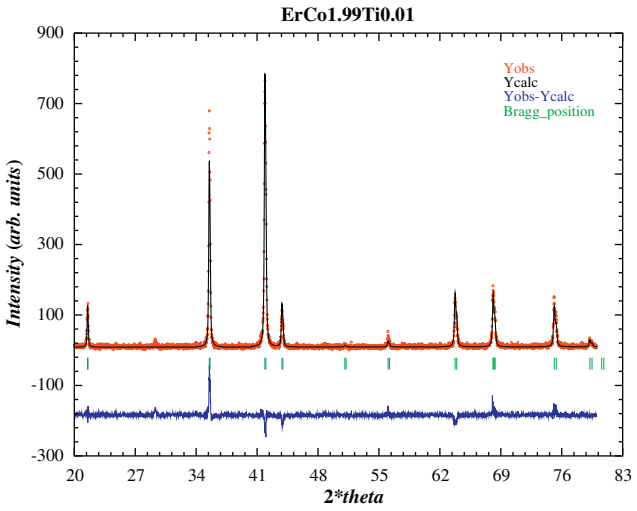

Fig. 1. X-ray powder pattern of $ErCo_{1.99}Ti_{0.01}$ with CuK_α radiation ($\lambda = 1.5418$ Å) at room temperature. The experimental data, the calculated profile and the difference between observed and calculated profiles are shown.

X-ray powder diffraction experiments were carried out using a Cu target source with the sample located on a zero background silicon carrier. Rietveld refinement confirmed that all compounds are mainly single phase, possessing the $MgCu_2$ type C-15 cubic Laves phase structure (space group $Fd\bar{3}m$). It was observed that, with the increase in Ti content (x), the relative intensity of the diffraction peaks clearly increased and the diffraction angles shifted very slightly to higher angles, up to the composition $x=0.04$, indicating a contraction of the unit cell. Fig. 1 shows a typical room temperature X-ray diffraction pattern, along with the Rietveld refined pattern, for $ErCo_{1.99}Ti_{0.01}$. However, an additional peak, though very weak maximum up to 5% of parent peak height, is also observed at $2\theta = 29.27_0$ for all the samples. According to our analysis, it does not belong to any binary Er–Co or Ti–Co phases. It is likely that this unidentified peak arises from some complex oxide (ErCoO) phases.

The lattice parameter decreases almost linearly with increasing x, from $a=7.1500$ Å for $ErCo_2$ to $a=7.1400$ Å for $ErCo_{1.96}Ti_{0.04}$, and then increases slightly to $a=7.1412$ Å for $ErCo_{1.95}Ti_{0.05}$. Further details including lattice parameters values can be found in [11]. Note that the Ti solubility in Er–Co–Ti in the concentration range of the samples studied is in very good agreement with the Er–Co–Ti system phase diagram measured very recently by Kotur et al. [12].

3. Results and discussion

3.1. Resistivity measurements

The temperature variations of the resistivity, ρ (T), normalized by the values at $T=273$ K, are plotted for the different samples in Fig. 2. A sharp drop is observed at a critical temperature (T_c)