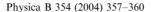


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# AC susceptibility in $Y_{1-x}Tb_xCo_2$ compounds

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#### Abstract

AC susceptibility measurements, as a function of temperature and frequency, have been carried out in  $Y_{1-x}Tb_xCo_2$  ( $0.1 \le x \le 0.9$ ) compounds to investigate the magnetic phase transition. It is known that  $YCo_2$  exhibits very strongly enhanced Pauli paramagnetism, while  $TbCo_2$  orders magnetically with a Co moment induced by the Tb molecular field. The phase transition of  $TbCo_2$  at 231 K is a second-order phase transition. We found that in  $Y_{1-x}Tb_xCo_2$  the transition changes from first- to second-order as x increases. The increase in the order temperature with Tb concentration is observed and the type of the phase transition is analyzed in the framework of the Inoue–Shimizu model. © 2004 Elsevier B.V. All rights reserved.

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Keywords: RCo2; Magnetic phase transitions; AC susceptibility

## 1. Introduction

 $Y_{1-x}R_xCo_2$  (R = rare earth) is a good candidate for studying the magnetism of 3d–4f intermetallic compounds. In YCo<sub>2</sub> both Y and Co bear no magnetic moment, the compound exhibits a very strongly enhanced Pauli paramagnetism and undergoes a first-order metamagnetic transition under an external field exceeding a certain critical

value. The addition of a heavy R, like Tb, provides a strong molecular field which induces a magnetic moment on the Co ( $\cong 1.0 \mu_B$ ) through the polarization of the 3d band. The induced Co moment aligns antiparallel to the Tb moment and the system orders ferrimagnetically [1]. The magnetic phase transition in RCo<sub>2</sub> is of second-order for R=Tb and Gd, of first-order for R=Dy, Er and Ho.

In this work we present results from AC susceptibility measurements on the pseudo-binary alloys  $Y_{1-x}Tb_xCo_2$ . These compounds have been previously studied using low-temperature specific heat [2], magnetization measurements [2,3], electric

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resistivity [2], thermal expansion [3], neutron diffraction [4], AC susceptibility [3,4] and NMR [5]. The aim of the present work is to study how the change in the order of the magnetic transition is reflected in the susceptibility.

## 2. Experimental

 $Y_{1-x}Tb_xCo_2$  pseudo-binary compounds with  $x=0.1,\,0.2,\,0.3,\,0.4,\,0.5,\,0.6,\,0.7,\,0.8$  and 0.9 were prepared by arc melting the stoichiometric mixture of the elements in an Ar atmosphere. To improve homogeneity, the buttons were crushed and melted several times. Samples were characterized with X-ray diffraction (XRD). The AC susceptibility measurements were performed in a Lake Shore 7130 susceptometer using a sample cup containing about 50 mg of each sample. To avoid nonlinear magnetization effects, a low enough field  $H_{AC}=1$  Oe was used. Data were acquired at frequencies ranging from 5 to 9920 Hz in a temperature range of 15–300 K.

#### 3. Results and discussions

According to the XRD measurements (see Fig. 1) all samples have the crystal structure of a cubic C15 Laves phase. The nonindexed diffraction peaks correspond to those of YCo<sub>3</sub> indicating a small contamination by this intermetallic compound. The origin of the reflection at  $2\theta = 29^{\circ}$  has not been identified.

Typical curves  $\chi'(T)$  of the real components of the susceptibility at different frequencies are shown in Fig. 2, for the samples with x = 0.3 and 0.9, respectively. As no clear frequency dependence has been observed we choose data obtained at 825 Hz in order to compare samples with different Tb concentration. Concentration dependences of  $\chi'(T)$  are shown in Fig. 3. As the Tb concentration increases, the peaks shift to higher temperatures evidencing an increase in the order temperature  $(T_c)$ . One also observes a change in the shape of the  $\chi'(T)$  curve: for  $x \le 0.4$  the curve is symmetric with respect to its maximum while for x > 0.4  $\chi'(T)$  is asymmetric (lambda form; see also

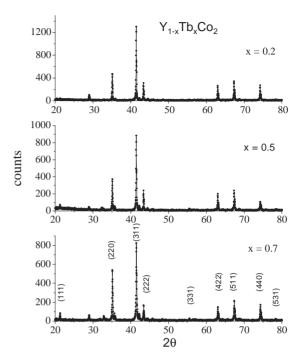


Fig. 1. Typical X-ray diffraction patterns. Indexed lines correspond to C15 structure.

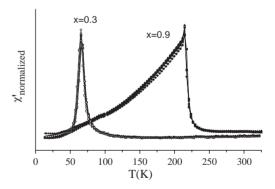


Fig. 2. Temperature dependence of real component of AC susceptibility recorded at various frequencies (5, 48, 375, 825, 3320 and 9920 Hz) for two samples (normalized data).

Fig. 2 for a clearer comparison of the two types of curves). The  $T_c$  values are plotted in Fig. 4 together with those from Ref. [3] and the value for TbCo<sub>2</sub> from Ref. [6]. Our values were obtained analyzing the thermal variation of the inverse paramagnetic susceptibility (see inset in Fig. 4).

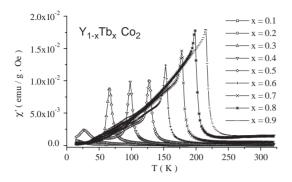


Fig. 3. Temperature dependence of real component of AC susceptibility recorded at 825 Hz for the whole range of concentrations.

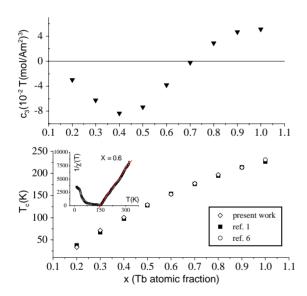


Fig. 4. Up: coefficient  $c_3$  taken from calculations of Ref. [3] as a function of  $T_{\rm c}$ . Down: order temperature  $T_{\rm c}$  as a function of Tb atomic fraction. Inset: typical inverse susceptibility vs. temperature, used to determine the  $T_{\rm c}$  value from the linear fit in the paramagnetic region.

Different peak shapes in  $\chi'(T)$  have been related in the literature to the order of the phase transition: a lambda-type is frequently taken as an indication of a second-order transition, a more symmetric peak is associated with a first-order transition [3]. In the case of  $\text{Er}_{1-x}\text{Tb}_x\text{Co}_2$ , a sharp drop of  $\chi'(T)$  just below  $T_c$  has been taken as a characteristic of first-order transition [7]. Theoretically, the change of order has been shown by Bloch et al. [1] and Inoue

et al. [8] to be related to the sign of the coefficient  $c_3$  of the  $M^4$  term in the Landau expansion of the free energy in powers of the total molar magnetic moment M. Using the experimentally determined  $T_{\rm c}$  values the coeficient  $c_3$  can be calculated [3]. As there is a good agreement between our  $T_c$  values and those from Ref. [3] we assumed those  $c_3$  values (see Fig. 4) to apply in our case also. The calculation predicts a change in the order of the transition near x = 0.7. Measurements of the magnetic hyperfine field at Cd impurities in  $Y_{1-x}Tb_xCo_2$  by means of the perturbed angular correlation technique, carried out on the samples used in the present work [9], indicate a first-order transition for  $x \le 0.7$  and a  $T_C$ -dependence of the discontinuous jump of the order parameter at the first-order transition.

Although a well-defined change in the form of  $\chi'(T)$  is observed (see Fig. 2), the comparison with the PAC data shows that the determination of the critical concentration for the change of order from  $\chi'(T)$  data only is a difficult task. Our susceptibility results are in agreement with a scenario of gradual change. The identification of the order of the transition in 3d–4f systems requires a better understanding of the relation between the characteristics of the phase transition and the shape of the response of the AC field.

#### 4. Conclusions

The temperature dependence of the magnetic response of  $Y_{1-x}Tb_xCo_2$  ( $0.2 \le x \le 0.9$ ) compounds was determined by AC susceptibility measurements. This study reveals that the transition change from first-order towards a second-order as x increases. The comparison with angular correlation data suggests that it is not possible to establish a critical concentration for the change of order of the transition from AC susceptibility data alone.

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