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To cite this article: D P Rojas *et al* 2011 *J. Phys.: Conf. Ser.* **325** 012023

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Antiferromagnetic behaviour of Tb₂Al alloy

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Abstract. The structural, thermal and magnetic properties of the Tb₂Al alloy have been investigated by AC/DC magnetic susceptibility, specific heat, X-ray and neutron diffraction measurements. DC and AC-magnetic susceptibility results are consistent with an AFM order at $T_N = 52$ K. The specific heat data show a lambda anomaly associated to the magnetic transition with a peak at 52 K ($c_{ord} = 99$ J/molTbK). The analysis of thermodiffraction patterns of neutron diffraction indicates that, below the ordering temperature, the magnetic reflections can be indexed with a commensurate lattice related to the crystallographic cell (*Pmna*) by a propagation vector $k = (1/2, 1/2, 1/2)$. The results are compared with those reported for other magnetic rare earth alloys of R₂Al-type (with R = Nd, Gd and Dy).

1. Introduction

Rare earth metals and alloys have been object of extensive studies because their interesting phenomena related to exotic magnetic structures, high magnetocrystalline anisotropy, crystal field effects and uncommon magnetic transitions [1, 2, 3]. Among them, some Tb-Al binary alloys have been particularly studied by neutron scattering techniques [4, 5, 6]. From those studies, it was concluded that TbAl₂ shows ferromagnetism at $T_C = 105$ K [4], TbAl is a non-collinear antiferromagnet with $T_N = 72$ K [5], whilst Tb₃Al₂ is a ferromagnet with $T_C = 190$ K [6]. On the other hand, studies of magnetic properties of other members of the Tb-Al series indicated an antiferromagnetic behaviour at $T_N = 21$ K and 52 K for TbAl₃ and Tb₂Al, respectively [1, 7, 8]. However, in some cases as the Tb₂Al alloy, it was evident that the magnetic properties can be affected by the presence of impurities [7, 8], and therefore, the influence of minor spurious magnetic contributions should be carefully investigated. For this reason, in the present work, we will provide a more detailed description of the magnetic properties of Tb₂Al alloy by DC and AC-susceptibility measurements, focusing on the frequency and field dependence at low magnetic fields, below 0.5 kOe. In addition, a complementary study with neutron diffraction measurements will also help to shed light on the magnetic state at the microscopic level.

2. Experimental details

A polycrystalline Tb₂Al alloy was prepared in an arc furnace from stoichiometric amounts of Tb (3N Alfa) and Al (5N, Alfa) metals. X-ray diffraction measurements were carried out in a Philips diffractometer with CuK_α radiation. AC (DC) magnetic susceptibility and specific heat measurements were collected in a Quantum Design PPMS in the temperature range 2 - 300 K

and magnetic fields up to 9 T. Neutron diffraction measurements were recorded at the G4.1 diffractometer ($\lambda = 2.426 \text{ \AA}$) of the Orphée reactor in the Leon Brillouin Laboratory (LLB) with a position sensitive detector of 800 cells, spanning an angular range of 80° (from 10° to 90°).

3. Results and Discussion

Rietveld refinements of X-ray powder diffraction experimental data are consistent with an orthorhombic Co_2Si -type structure, space group $Pnma$, with two inequivalent crystallographic positions for the Tb atoms, and lattice parameters $a = 6.564(1) \text{ \AA}$, $b = 5.118(1) \text{ \AA}$ and $c = 9.449(2) \text{ \AA}$, in good agreement with those reported in the literature [7, 8]. In addition, the diffractogram does not reveal the presence of extra peaks associated to impurity phases. However, according to the results of Tb-Al phase diagram, Tb_2Al was indicated to form peritectically [9], and therefore, the presence of other binary Tb-Al alloys, below the detection limit in X-ray diffraction, should not be discarded.

The temperature dependence of zero field cooled (ZFC) and field cooled (FC) DC-magnetic susceptibility ($\chi = M/H$) curves at different magnetic fields is shown in Figure 1. At the magnetic field of 0.02 kOe, four anomalies can be detected: a peak at $T_N = 52 \text{ K}$, associated to the AFM ordering [7, 8], and three additional ones around T_{max} , T^* and T^{**} . These last magnetic contributions broaden with the increase of the magnetic field and practically disappear for 0.5 kOe, where the ZFC and FC curves coincide, thus indicating, that their origin could come from impurity phases. On the other hand, the field dependence of the isothermal magnetization at 15 K (see Figure 2), shows a typical AFM (linear) behaviour to the highest applied magnetic field in our experiment, whereas the curve at 48 K displays a similar behaviour up to 60 kOe, field at which a metamagnetic transition takes place.

Measurements of the AC-susceptibility also provide information about the origin of the low magnetic field contributions. In Figure 3, the temperature dependence of the real (χ') and imaginary (χ'') components of the AC-susceptibility at several frequencies is presented. Similarly to the results of the DC-susceptibility at the low magnetic field of 20 Oe, four anomalies at T_N , T_{max} , T^* and T^{**} are found. As commented above, the magnetic contributions at T_{max} , T^* and T^{**} may be ascribed to impurity phases. It is known, for instance, that the binary TbAl alloy orders around 72 K [5], which coincides with the position of the anomaly at T^* . Concerning the other two anomalies, they can be attributed to the contribution of Laves phase alloy TbAl_2 with a ferromagnetic ordering around 100 K (T^{**}) [4, 10], and with a dynamic domain wall

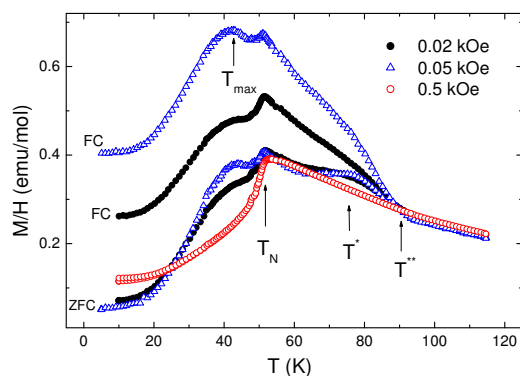


Figure 1. Temperature dependence of ZFC-FC curves of DC-magnetic susceptibility at different magnetic fields.

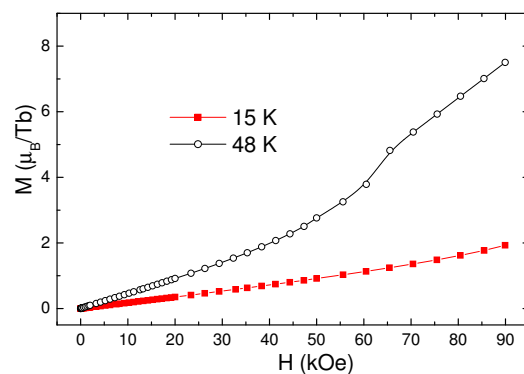


Figure 2. Field dependence of the isothermal magnetization curves at 15 and 48 K for Tb_2Al alloy.

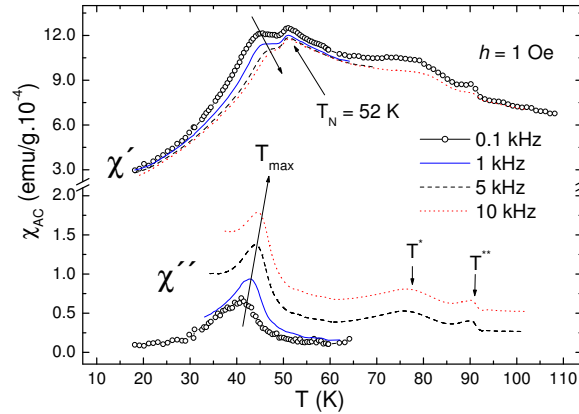


Figure 3. Temperature dependence of the real and imaginary components of AC-susceptibility of Tb₂Al alloy at different frequencies.

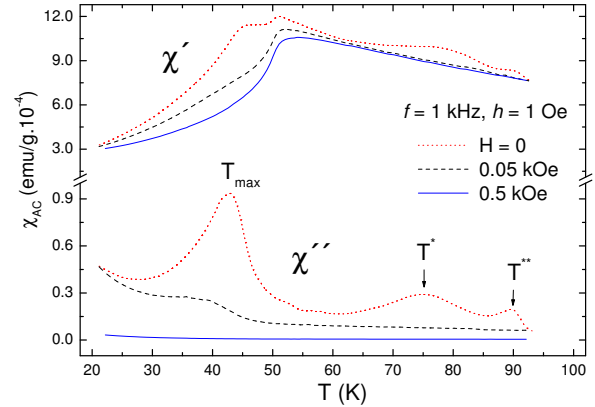


Figure 4. Field dependence of the $\chi'(T)$ and $\chi''(T)$ at the frequency of 1 kHz and AC magnetic field $h = 1$ Oe.

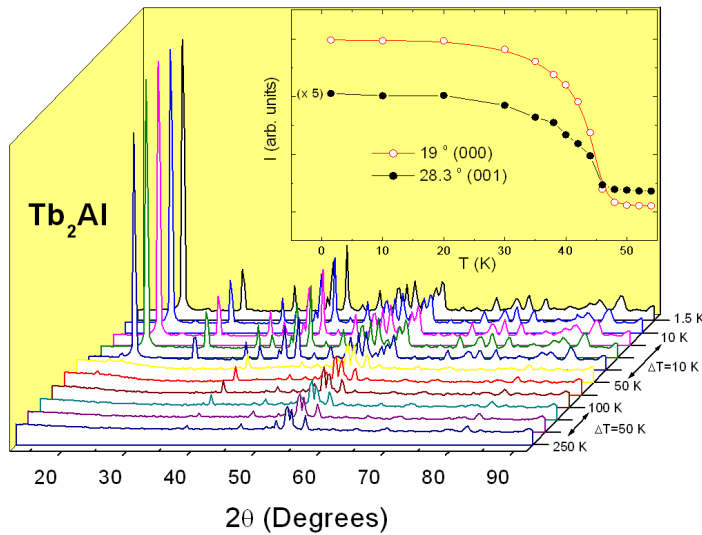


Figure 5. Thermodiffractograms of neutron diffraction patterns of the Tb₂Al alloy at different temperatures. The inset details the temperature dependence of the two most intense magnetic reflections.

thermal activation process which takes place around 45 K, being described by an Arrhenius relaxation law with $\tau = 1.3 \cdot 10^{-11}$ s and $E_a = 803$ K (69 meV) [10]. The contributions of these impurity phases are masked by the magnetic field, as shown in figure 1. A similar effect is also observed from the field dependence of the AC-susceptibility (see Figure 4), where for a biasing magnetic field of 0.5 kOe, only the expected AFM contribution at $T_N = 52$ K of the main phase (Tb₂Al alloy) is found. In other recent studies on Tb₂Al alloy, and additional ferromagnetic contribution around 235 K, probably associated to Tb metal crystallites, was evidenced [8], which is not observed in our case. This confirms the difficulty to obtain pure alloys.

Specific heat is also an useful tool in the study of magnetic transitions. From the results of measurements on the Tb₂Al alloy (not shown), we have found a lambda anomaly with a peak at 52 K ($c_{ord}=99$ J/molTbK), associated to the AFM transition. It is worth commenting that no additional anomalies were observed, specially at the temperatures T_{max} , T^* and T^{**} , where the impurity contributions in the DC and AC-susceptibility were found.

In figure 5, thermodiffractograms of neutron diffraction patterns of the Tb_2Al alloy at different temperatures are presented. It is observed that new peaks appear at low angle positions between 15° and 30° , when the temperature decreases from 52 K and down to 1.5 K. These peaks do not correspond to the crystallographic reflections expected in the $Pmna$ space group, hence corresponding to an AFM type of magnetic structure. The temperature evolution of these magnetic Bragg peaks allows to estimate the ordering temperature, in good agreement with DC and AC-susceptibility results, as observed from the thermal variation of the integrated magnetic intensity of the two strongest magnetic reflections (000/010), displayed in the inset of Figure 5. Even though we have not succeeded in the determination of the full magnetic structure, from the analysis of the patterns below $T_N = 52$ K, we have found that all the observed Bragg peaks can be indexed with a commensurate lattice related to the crystallographic cell ($Pmna$) by a propagation vector $k = (1/2, 1/2, 1/2)$ (the components are referred to the reciprocal vectors of the conventional unit cell).

The careful analysis of the magnetic contributions from TbAl and TbAl_2 is consequently useful to evaluate their influence in the overall behaviour of the studied alloy. In particular, the AFM character of Tb_2Al alloy is not masked and is perfectly recorded in the neutron diffraction patterns, where the peaks associated to impurity phases are hardly observed.

If we compare this with the results reported for other R_2Al alloys, it was found that Nd_2Al is ferromagnetic with an ordering temperature of 36 K, whereas Gd_2Al and Dy_2Al order AFM with $T_N = 46$ K [8] (close to the transition observed in Tb_2Al alloy) and 37 K [7], respectively. The magnetic field dependence of the magnetization in this type of compounds suggests the presence of a high magnetocrystalline anisotropy, and in some cases of metamagnetic transitions [7, 8], as obtained here in the case of the Tb_2Al alloy (see Figure 2).

4. Conclusions

The results of DC(AC)-susceptibility on the Tb_2Al alloy are consistent with an AFM behaviour at $T_N = 52$ K. This is also supported by the analysis of thermodiffractograms of neutron diffraction patterns in good agreement with an AFM structure. It would be interesting to extend this study to other R_2Al systems with $\text{R} = 4f$ elements, crystallizing in the hexagonal Co_2Si structure.

Acknowledgments

This work was supported by the Ministry of Science and Innovation of Spain under Contract No. MAT 2008-06542-c04-04.

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