Local magnetic properties of (RE)$_{12}$Co$_5$Bi studied by $\mu$SR

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Abstract

The recently discovered (RE)$_{12}$Co$_5$Bi (where RE is a rare earth element) system is of great scientific interest due to its rich magnetic phase diagram. In this paper, we report studies on polycrystalline samples of RE = Tb, Gd and Tm. The RE=Tm compound is particularly interesting, with strong evidence for meta-magnetic behavior.

Key words: $\mu$SR, Rare earth intermetallic compounds, local magnetism

1. Introduction

Over the decades there has been enormous interest on rare-earth oxides[1–5] and intermetallic compounds[6,7] due to their rich phase diagrams. In particular, coupling of the f and d electrons in rare earth transition metal intermetallics give rise to many instances of complex magnetic phase diagrams and intriguing unresolved magnetic and transport phenomena. One such example is the newly discovered series of (RE)$_{12}$Co$_5$Bi where RE is a rare earth element. In this system, Bi occupies certain sites leading to a highly ordered structure with negligible quenched disorder due to its size.[6] Bulk magnetic studies suggest a variety of magnetic phases including ferrimagnetism and antiferromagnetism.[6] As usual, it is useful to complement the bulk magnetization studies with a local probe of magnetism such as $\mu$SR. MacFarlane et al.[8] have carried out such studies on the Ho$_{12}$Co$_5$Bi compound. These measurements revealed a bulk transition at $T_c = 30$ K to a magnetically ordered state, and it was shown that in the paramagnetic region, the sample follows a Curie-Weiss law with slow magnetic fluctuations which persist far above $T_c$. Keeping in mind the fact that magnetic properties in (RE)$_{12}$Co$_5$Bi could be dramatically modified by substitution of different RE atoms, it is also be interesting to study other members of the series.

In this paper, we report studies of the local magnetism in polycrystalline samples of selected (RE)$_{12}$Co$_5$Bi compounds for RE = Tb, Gd and Tm. Our results indicate magnetic transitions in the RE=Tb and Gd samples at around 75 K and 100 K respectively. On the other hand, the RE=Tm sample does not show any magnetic ordering down to 3 K in low magnetic fields. However, the effects of high magnetic field on the $\mu$SR spectra strongly suggests the existence of a meta-magnetic transition in Tm$_{12}$Co$_5$Bi where the magnetic ground state in low applied longitudinal magnetic fields is dramatically different from the one at high fields.

2. Experimental Details

The samples of Tb$_{12}$Co$_5$Bi, Gd$_{12}$Co$_5$Bi and Tm$_{12}$Co$_5$Bi were prepared by arc-melting of cold-pressed pellets prepared from stoichiometric amounts of elemental components with an excess of 2.5 wt % Bismuth added to compensate for vaporization during the reaction. The reaction was carried out in an Edmund Buhler MAM-I compact arc furnace on a water-cooled copper plate in a Ti-gettered argon atmosphere. Afterward, the resulting ingots were annealed in sealed evacuated silica tubes at 870 K for at least one week and then quenched in cold water. All the sam-
samples were characterized by powder X-ray diffraction on an Inel powder diffractometer equipped with a CPS 120 detector and found to be single phase. Further details of sample preparation and structural characterization can be found elsewhere.[6] The magnetization measurements were carried out using a Quantum Design PPMS system.

The µSR measurements were carried out on the M20B beamline at TRIUMF in Vancouver, Canada, where µSR and 3NMR experiments can be carried out.[10–12] In this experiment, a beam of nearly 100% spin-polarized positive muons (lifetime 2.2 μs) of nominal momentum ~28 MeV/c is implanted into the sample. The sample was mounted in a packet made from high-purity Ag foil (thickness 25 μm). The packet was attached to a high-purity silver backing with Apiezon N grease in a helium gas flow cryostat. The experiments described in this paper were carried out in the standard longitudinal field (LF-µSR) configuration.[9,13,14] A small longitudinal field (LF) of ~1 mT was applied to ?decouple? relaxation effects due to nuclear moments (¹⁴RE, ⁵⁸Co, and ⁸⁹⁹Bi, where A is the atomic number) and any small stray magnetic fields, and hence we are only sensitive to the stronger electronic moments.[12] A signal that relaxes or oscillates faster than the deadtime of the spectrometer (~10 ns) is detected only as a missing fraction of the total asymmetry.[8]

3. Results and Discussion

Fig. 1 summarizes the temperature dependence of the bulk DC susceptibility χ and inverse susceptibility of the three samples, measured in a field of 0.5 Tesla. These results are discussed in more detail in Ref. [6] and we only briefly summarize them here. There is a peak in the χ(T) of Tb₁₂Co₅Bi at approximately 75 K this is believed to be a consequence of an antiferromagnetic transition. Above this temperature, the Tb₁₂Co₅Bi sample is in the paramagnetic state. In the case of Gd₁₂Co₂Bi, the susceptibility rises rapidly below 120 K. By also taking into account the magnetic field dependence of the DC and AC susceptibility data, it was suggested that the sharp rise is a result of ferrimagnetic ordering in this sample.[6] The Tm₁₂Co₅Bi appears to be in a paramagnetic state down to 3 K (see Fig.1), but there may be an antiferromagnetic transition at Tₐ ≈ 2.7 K.

Let us now discuss our µSR results on these samples. In all three compounds, no coherent precession signals were observed at any temperature, similar to the situation in Ho₁₂Co₅Bi reported previously by MacFarlane et al.[8] In Figure 2, we show representative LF-µSR spectra in Tb₁₂Co₅Bi. The data are well-described by a single component exponential relaxation together with a non-relaxing component (most likely due to muons that stop in the surrounding Ag):

\[ A(t) = A_1 e^{-λt} + A_Ag \]  

For fitting the LF = 10 G data to Eq.(1) we were able to extract the spin relaxation rate λ and the amplitude A₁ for Tb₁₂Co₅Bi and Gd₁₂Co₂Bi, clear evidence for a magnetic transition is taking place. As usual, the high temperature side corresponds to a paramagnetic state, while below the peak, there is static magnetic order. Consistent with this, A₁ decreases dramatically at the transition temperature Tc ≈ 75 K and 100 K for Tb and Gd (vertical line in Fig. 3 (a) and (b)), confirming that the majority of the sample has entered a magnetically ordered state. Note that these transition temperatures are roughly consistent with the bulk magnetization measurements.[6] On the other hand, there is no evidence in the Tm₁₂Co₅Bi sample for a magnetic transition within the temperature range of 3-300 K. According to the DC susceptibility measurements for the RE=Tm sample (in a field of 0.5 Tesla), antiferromagnetic ordering takes place at temperatures lower than 3 K, outside our accessible temperature range.

The relaxation rates at high temperatures, i.e. in the paramagnetic state, for the RE=Gd sample are substantially lower than for the RE=Tb and Tm samples. Similar behavior has been observed in rare-earth Gd and Tb...
quasicrystals.[15] We speculate that the same qualitative mechanism proposed there might also explain our results: In the fast fluctuation regime, the observed muon spin relaxation rate is determined by the ratio of the second moment of the field distribution at the muon site, $\langle B^2 \rangle$, and the field fluctuation rate. If the site is assumed to be the same in the three compounds, then $\langle B^2 \rangle$ is determined by the magnitude of the surrounding moments. The free-ion moments for Tb$^{3+}$, Tm$^{3+}$ and Gd$^{3+}$ are comparable, so the large differences in relaxation rates may indicate that the Tb and Tm moment fluctuation rates are much less than in Gd. These differences in the fluctuation rates may be a consequence of crystalline electric field (CEF) splittings originating from the low rare earth site symmetry. Note that for Gd$_{12}$Co$_5$Bi, the trivalent Gd is in a state of zero total angular momentum.[14] In this case the ion is spherically symmetric so it is not affected by the CEF.[14–16] However, CEF effects would be important in Tb$_{12}$Co$_5$Bi and Tm$_{12}$Co$_5$Bi. Measurements such as inelastic neutron scattering, heat capacity, etc. may be useful for elucidating the nature of the crystal field splittings, but are not currently available for these materials.

We also performed LF-$\mu$SR measurements in a magnetic field of 5 Tesla. In Figure 4, we compare the data at 10 Gauss with the 5 Tesla data for Gd$_{12}$Co$_5$Bi and Tm$_{12}$Co$_5$Bi. In Gd$_{12}$Co$_5$Bi, the magnetic transition temperature at 10 G and 5 T are the same (~100K). Furthermore, the relaxation rate in high fields is smaller than at low fields, the standard well-known behavior associated with probing the spectral density of magnetic fluctuations at a constant temperature.[9,14] As mentioned above, the low field magnetic transition temperature extracted from the LF-$\mu$SR measurement is consistent with the bulk magnetization results measured in a field of 0.5 Tesla.

By contrast, in Tm$_{12}$Co$_5$Bi, the relaxation rates at a particular temperature increases with increasing field (see Fig. 4b). This is clear indication that the magnetic states in low fields and high fields are different. Bulk magnetization studies on this sample also led Tkachuk et al.[6] to propose the existence of a metamagnetic transition above a field of 2 Tesla at 2 K, believed to originate from the collapse/melting of the antiferromagnetic ground state with applied field. It will be fruitful to investigate the low tem-

![Fig. 2. Typical raw data of the time dependence of LF Relaxation in a field of 10 Gauss for Re=Tb sample. The solid curves are best fits to Eq.(1).](image)

![Fig. 3. (a)-(b)-(c) Temperature dependence of relaxation rate $\lambda$ in a longitudinal field of 10 Gauss for RE=Tb, Gd and Tm. Insets show the corresponding relaxing asymmetry for each sample. The solid curves are guide to the eye.](image)
Fig. 4. (a)-(b) Temperature dependence of relaxation rate $\lambda$ in longitudinal fields of 10 Gauss and 5 tesla for RE= Gd and Tm. The solid curves are guide to the eye.

Temperature behavior of the RE=Tm sample using $\mu$SR. There are several questions regarding the observed unusual behavior: will the magnetic ordering temperature change with increasing magnetic field? What is the threshold field for the proposed metamagnetic transition?

Acknowledgements

This research is partially supported by the Center for Materials and Molecular Research at TRIUMF and the NSERC. TRIUMF receives federal funding via a contribution through the NSERC.

References


