Experimental report

09/02/2017

Proposal: **4-04-486** **Council:** 10/2016

Title: CRYSTAL FIELD EXCITATIONS IN BULK TbCu2, AS THE PRECURSOR METALLIC ALLOY OF SUPERANTIFERROMAGNETIC NANOPARTICLES

Research area: Physics

This proposal is a continuation of 4-05-626

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Samples: TbCu2
Tb0.1Y0.9Cu2

Instrument	Requested days	Allocated days	From	To
IN4	6	4	23/01/2017	27/01/2017

Abstract:

The energy structure and temperature dependence of the magnetic excitation spectra of bulk TbCu2 will be studied by inelastic neutron scattering, as the precursor of Superantiferromagnetic nanoparticles. The measurements will be performed on polycrystalline samples of TbCu2 and its analogue Tb0.1Y0.9Cu2 on the high intensity time-of-flight spectrometer IN4 in the temperature range 1.5 K $< T <$ 150 K. The crystal field (CF) parameters will be calculated and the CF splitting scheme of the 4f ground multiplet of Tb3+ ions will be also determined from the analysis of the INS spectra. The temperature dependence of these excitations is of relevance to make a calculation of the spin-wave spectrum of TbCu2 and nanostructured related materials.

ILL Experimental Report – Proposal 4-04-486

CRYSTAL FIELD EXCITATIONS IN BULK $TbCu_2$, AS THE PRECURSOR METALLIC ALLOY OF SUPERANTIFERROMAGNETIC NANOPARTICLES

Principal Investigator: Luis Fernández Barquín, Universidad de Cantabria, Spain
Instrument: IN4
Dates: 23/01/17 – 27/01/17

The attraction of Superantiferromagnetic ensembles of $TbCu_2$ magnetic nanoparticles (MNPs) is supported by the presence of an antiferromagnetic (AFM) core and a disordered magnetic shell whose magnetic coupling is relatively unexplored. To understand this coupling is a must if we were to apply correctly MNPs in biomedicine and waste management.

The collective moment dynamics is also far from being well-established in MNPs, and the scarce studies are concentrated on Fe-oxides, with troublesome and unstable magnetic structures. $TbCu_2$ is a much better defined system of MNPs and thus, a more feasible alloy to analyse the magnetic excitations. Thus, we conducted a preliminary experiment at IN6 with D = 12 & 9 nm nanoparticles (and a bulk control alloy) to show the changes between bulk and nanostructured dynamics.

It was then immediately obvious we needed to define the crystal field excitations in the paramagnetic regime and this had to be carried out in bulk $TbCu_2$. A diluted $(Tb_{0.1}Y_{0.9})Cu_2$ alloy permits a clearer definition of crystal field levels and hence it was included as well. In January 2017 we have performed the experiment at IN4; this successful experiment has been carried out as follows: The spectra were recorded at T= 1.5, 8, 20, 40, 100, 250 K. In $(Tb_{0.1}Y_{0.9})Cu_2$ the magnetic transition temperature is 5 K, as obtained in DC-susceptibility measurements. In $TbCu_2$,T_N = 50 K. To vary the incident energy λ = 1.1, 2.2 and 3.06 Å were employed allowing to cover a sufficiently large energy transfer interval. The data treatment is already performed including detector correction, elastic peak definition, background analysis and normalisation at the different temperatures.

One straightforward result is that we are getting excellent quality spectra with around 13 g of powder sample. In $(Tb_{0.1}Y_{0.9})Cu_2$ it is observed that there exist magnetic excitations (Q = 1.6 $Å^{-1}$) peaking at energy transfer values of 5.3 meV at T = 1.5 K (magnetic state). Clearly this is reduced when increasing temperature and another excitation appears peaking around 4 meV at T =100 K. (Fig. 1). There are expectations of 8 multiplets with evaluated crystal field parameters B_2^0 and B_2^2, obtained from the paramagnetic susceptibility (and simulations) [1].

In TbCu2 the main peak is observed at 7.7 meV (Q = 1.75 $Å^{-1}$), with a lower energy shoulder (Fig. 2). These become modified when T = 100 K (paramagnetic regime). There, a peak at 4.8 meV is the main contribution to the INS spectra (Fig. 2).]. We are now processing the data and calculations will be restarted at the University of Oviedo soon. Preliminary calculations with a main contribution around 5 meV are shown below (Fig. 3) in bulk TbCu2 need to be refined with the aid of new data at hand. Clearly, we are now in the position to perform a new (and concluding) experiment at IN4 in a new proposal, this time for the nanometric samples.

[1] M. Andrecut *et al.,* J. Phys. D **26**, 1144 (1993).

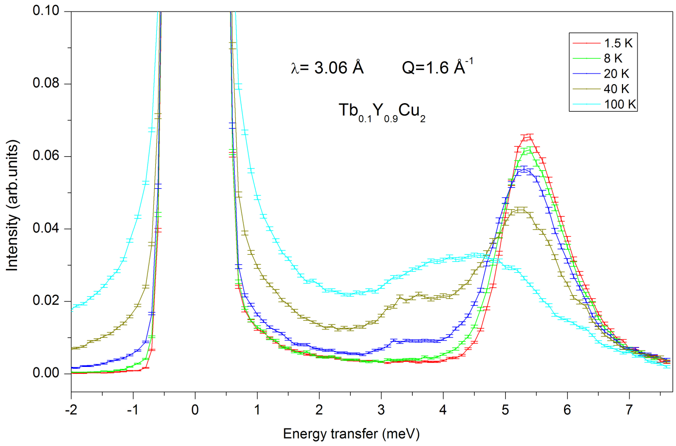

Fig.1. Inelastic neutron scattering data (IN4) measured at T = 100, 40, 20, 8 and 1.5 K for $(Tb_{0.1}Y_{0.9})Cu_2$ bulk sample at Q = 1.3 $Å^{-1}$.

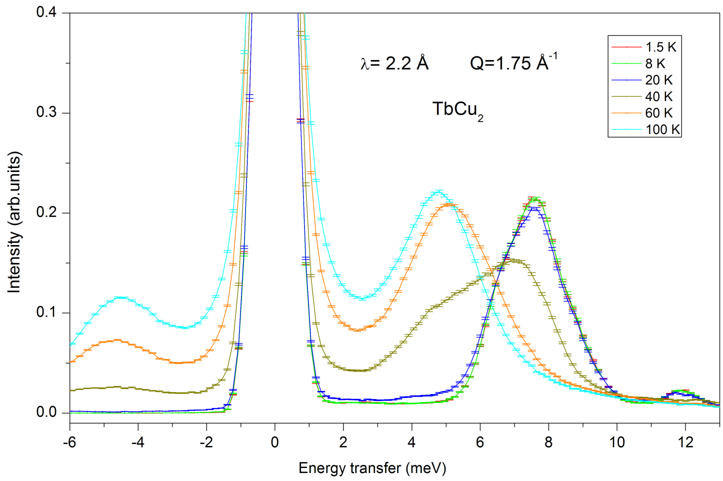

Fig. 2. Inelastic neutron scattering data measured of $TbCu_2$ bulk sample (IN4) at T = 100, 60, 40, 20, 8 and 1.5 K and at Q = 1.75 $Å^{-1}$.

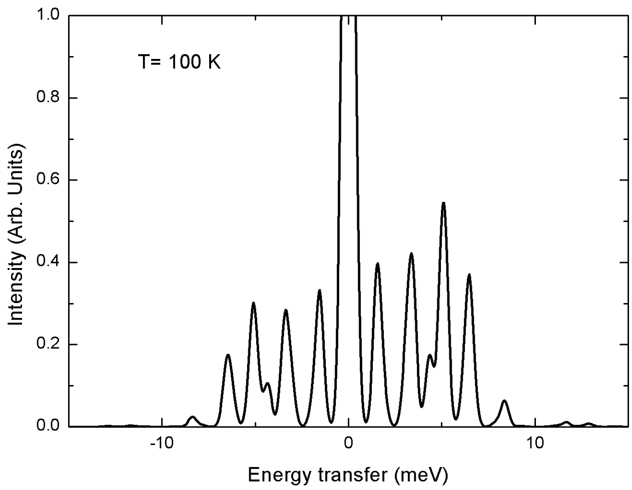

Fig. 3. Theoretical INS spectra in the paramagnetic state (T = 100 K) for $TbCu_2$ calculated with incident neutron energy of 15 meV. Our new experimental INS data will serve to refine such a model.