## **Experimental report**

Proposal:	4-05-626			<b>Council:</b> 4/2015			
Title:	Spin w	Spin wave spectrum in superantiferromagnetic TbCu2 nanoparticles					
Research area: Physics							
This proposal is a new proposal							
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Samples: TbCu2							
Instrument			Requested days	Allocated days	From	То	
IN6			0	3	01/12/2015	04/12/2015	
IN5			3	0			
Abstract:							

Superantiferromagnetic ensembles of TbCu2 magnetic nanoparticles constitute a case example of supermagnetic states, which effectively constitute barely unexplored magnetic arrangements. These TbCu2 nanoparticle ensembles present an antiferromagnetic core and a disordered moment shell due to the size reduction and the microstrain increase. In these ensembles, a pristine size-dependence of Neel and freezing transitions is found. Inevitably modifications of the spin wave spectrum are expected and very little is known regarding this fundamental issue. It is our aim to extract the dispersion curves. Clearly, the understanding of magnetic behaviour among nanoparticles is key to achieve new deliveries in present technological output (Horizon2020), especially in biomedicine and environmental conservation.

## ILL Experimental Report – Proposal 4-05-626

## "Spin wave spectrum in superantiferromagnetic TbCu2 nanoparticles"

Principal Investigator: Luis Fernández Barquín, Universidad de Cantabria, Spain Instrument: IN6 Dates: 01/12/15 – 04/12/15

Superantiferromagnetic ensembles of TbCu<sub>2</sub> magnetic nanoparticles (MNPs) constitute a case example of supermagnetic states, which effectively constitute barely unexplored magnetic arrangements. In recent years, we have extensively studied MNPs of TbCu<sub>2</sub>-based alloys, in which have been reported the existence of a new superantiferromagnetic state (SAFM). These MNPs retain the antiferromagnetic (AFM) structure of the bulk counterparts, with a Néel temperature around  $T_N = 50$  K, but also allow the presence of the randomly frozen (SG, spin glass) surface moments, with a freezing temperature around  $T_f = 20$  K. These MNPs are produced by ball milling in Ar-sealed WC containers at different times (t), and we are able to produce large quantities (grams).

The size-dependence of Néel and freezing transitions in this system entail modifications of the spin wave spectrum and which are not well known. It is our aim to extract the dispersion curves in order to understand the dynamic magnetic behaviour of the TbCu<sub>2</sub> nanoparticles. For such a purpose, the INS experiment was carried out on three samples of TbCu<sub>2</sub> with different average particle size. These samples were: TbCu<sub>2</sub> Bulk, TbCu<sub>2</sub> milled 2 hours, with an average size of 12 nm, and TbCu<sub>2</sub> milled 5 hours, with an average size of 9 nm. During the inelastic neutron experiment, we could measure at different temperatures, 100, 40, 20, 8 and 1.8 K with an incident wavelength of 5.1 Å.

After performing the experiment, the proper data treatment was carried out at the ILL by Maria de la Fuente under the supervision of Björn Fåk. This was supported by a short stay grant (2 months) of the Spanish Ministry of Economy and Competitiveness. (Grant reference: EEBB-I-16-10921). The treatment included detector correction, elastic peak definition, background analysis and normalisation for the three samples of TbCu<sub>2</sub> measured at the different temperatures.



Fig.1. Inelastic neutron scattering data measured at 100, 40, 20, 8 and 1.8 K for the TbCu<sub>2</sub> bulk sample at different Q values;  $Q = 0.6 \text{ Å}^{-1}$  (on the left top),  $Q = 1.3 \text{ Å}^{-1}$  (on the right top),  $Q = 1.1 \text{ Å}^{-1}$  (on the bottom, zoom on the right).



Fig.2. Inelastic neutron scattering data measured at 100, 40, 20 and 1.8 K for the TbCu<sub>2</sub> 2 hours milled sample at Q = 1.1 Å<sup>-1</sup> (zoom on the right).



Fig.3. Inelastic neutron scattering data measured at 100, 40, 20 and 1.8 K for the TbCu<sub>2</sub> 5 hours milled sample at  $Q = 1.1 \text{ Å}^{-1}$  (zoom on the right).

In view of these results, we observed an anti-Stokes peak around an energy transfer of - 5 meV due to a magnetic contribution of crystal- field excitations. We can see variations on the intensity of the peaks and also of the width of the elastic peak.



Fig.4. Lorentzian half-width  $\Gamma$  of the elastic peak for each sample of TbCu<sub>2</sub> as a function of temperature.

In Figure 4, we can see the variation of the width of the elastic peak for each temperature. The three samples follow the expected response for crystal-field excitations in according to E.A. Goremychkin, *et al.* in Nature Physics, **4** (2008) 766. Although the changes for nanocrystalline state are evident, a quantitative definition of the CEF is pursued in this investigation. We are already calculating the levels in the spectra and Gaussian contributions have been included to explain the elastic peak and another one is used for the anti-Stokes peak. Comparisons to NdCu<sub>2</sub> and ErCu<sub>2</sub> bulk alloys show that it is a must to define precisely the CEF levels. In consequence we anticipate that new INS data on bulk samples will be necessary.