Experimental report

09/02/2016

Proposal: **5-41-842**

Council: 4/2015

Title: Magnetic structure analysis of quasicrystal approximants in the Au-Si-Tb system, by single crystal neutron diffraction.

Research area: Materials

This proposal is a new proposal

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Samples: $Au5Si1.3Tb$

Instrument	Requested days	Allocated days	From	To
D10	14	5	25/11/2015	30/11/2015

Abstract:

Since the discovery of the first icosahedral quasicrystal (QC) by Shechtman et al., their detailed atomic structures eluded researchers for many years until the recent structure solution of the binary icosahedral Cd5.7Yb quasicrystal. As far as the magnetism of moment bearing QCs, no long-range magnetic order but only spin-glass like freezing has been observed to date, and hence, the spin-glass behavior has been regarded even as an intrinsic property of magnetic clusters with icosahedral symmetry. However, it was recently shown that their crystalline counterparts, i.e., approximant crystals (ACs), exhibit magnetic transitions. Recently we reported observations of ferromagnetic (FM) transitions in Au-based approximants Au-SM-RE (SM=Si,Ge), which are ternary alloys related to the Cd5.7Yb quasicrystal, and are composed of so-called Tsai-type clusters containing RE12 icosahedra. Our present aim is to perform the first detailed investigations of magnetic structures on these quasicrystal approximants by single crystal neutron diffraction, in order to understand the relation between the structural variations and the magnetic properties.

Experimental Report 5-41-842
Magnetic structure analysis of a quasicrystal approximant in the Tb-Au-Si system by single crystal neutron diffraction

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Recently, a new class of magnetic materials: quasicrystal approximants (ACs) have emerged[1,2]. These materials were known mainly due to their structural, physical property and compositional similarities with quasicrystals (QCs)[3]. However, unlike QCs which are non-periodic[4,5] and hence does not allow standard approaches to determine their atomic and magnetic structures, ACs are conventional crystals with 3D periodicity and similar local structure to their corresponding QCs[6]. Therefore, the magnetic behaviors of ACs have been investigated alongside the QCs for more than three decades[7]. Nevertheless, all the investigations had provided only localized moments, short range magnetic orders and spin glass behaviors for both the QCs and ACs. The story has changed for the ACs in 2010 when Tamura et. al. reported the first long range anti-ferromagnetic ordering in a $TbCd_6$ AC[1]. However, for the QCs besides the tremendous and lengthy endeavor, there is no experimental report of long range magnetic ordering till to date.

Finding long range ordered magnetic QCs could provide unique magnetic behavior that would trigger distinct scientific and commercial interest. Long range magnetic order is a consequence of local interaction between magnetic moments. Hence, understanding the local microscopic magnetic structure of QCs could give an insight in to how magnetic moments interact, which could be an invaluable input in order to chemically design a QC to be magnetically long range ordered. The local magnetic structure of QCs can be obtained from microscopic study of magnetic ACs. This, however; is relatively new spectrum of study since the first magnetic structure refinement of an AC was reported in 2014 for a Tb-Au-Si ACs by us[8].

The reported magnetic structure of the Tb-Au-Si AC was determined from powder neutron diffraction data on the WISH instrument at ISIS facility. The magnetic structure was described as 'ferrimagnetic-like' and unprecedented different magnetic moment values on symmetrically equivalent Tb ions on the nuclear structure were obtained. This result invited further investigation of the magnetic structure of the Tb-Au-Si ACs rather on a single crystal. Hence, single crystal of Tb-Au-Si AC was grown by self-flux syntheses method using Au-Si alloy as a flux for the purpose of this experiment.

The magnetic property of a Tb-Au-Si high quality single crystal of 8 mm^3 was first studied by a Quantum design SQUID magnetometer. Figure 1 (a) and (b) shows the zero field cooled (ZFC) and field cooled (FC) magnetization as a function of temperature (M vs T) in an applied field of 800 A/m for the Tb-Au-Si AC in plane and out of plane to the (100) direction, respectively. This is more clearly seen in the insets of figure 1 (a) and (b) which depict the derivative of the FC curves (dMFC/dT) and the maximum slope is taken as

T_c. It can be seen that the magnetic transition temperature T_c is 11.5 K. Measurements of magnetization as a function of magnetic field (M vs H) were made to determine the saturation magnetization and coercivity of the sample in plane and out of plane to the (100) crystallographic direction. The results are shown in figure 1 (c) which indicates a clear magneto-anisotropic property in the different directions.

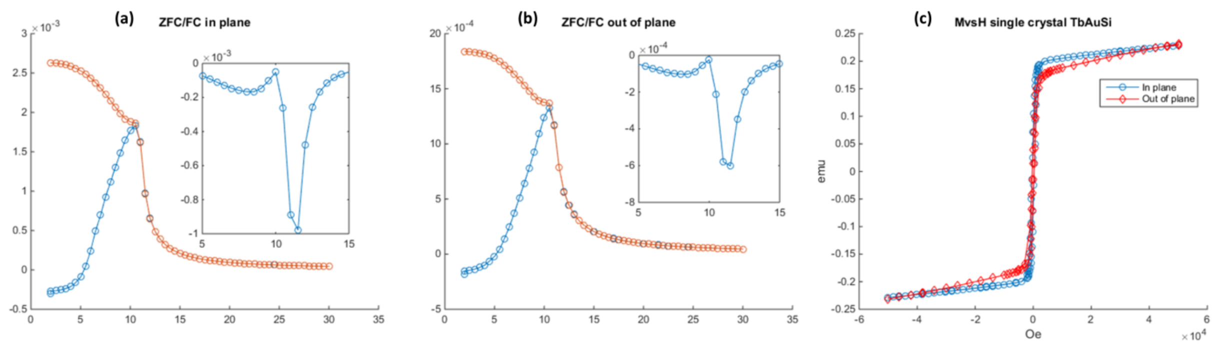

Figure 1: Magnetization vs temperature graphs for Tb-Au-Si AC (a) in plane to (100) direction, (b) out of plane to (100) direction; the insets in each plot are derivatives of FC curves vs temperature. (c) Plots of magnetization vs applied magnetic field (M vs H).

A high quality single crystal of 8 mm^3 was used for single crystal neutron diffraction (SCND) measurement at ILL facility of the D10 instrument in four-cycle configuration with a wavelength of 2.36 Å. The crystal was first aligned along the 2-fold direction with the help of a Laue neutron diffraction set up. We referred to the calculated intense peaks generated using the structure model obtained from single crystal x-ray diffraction (SCXRD) refinement on a single crystal of 10^{-3} mm^3 sliced from the current grain. Then narrow and short scans around the calculated strong nuclear peaks were measure at room temperature in order to obtain the orientation matrix. Using the obtained orientation matrix the alignment of the crystal was refined. The positions of the strong nuclear peaks were centered. The sample was then cooled to 20 K above the magnetic transition temperature (T_c) and the orientation matrix was realigned to compensate for the shrinkage of the unit cell and the misalignment of the crystal from the beam center (refined positions were < 0.05 ˚).

An actual data collection was made around selected 131 peaks (120 strong peaks and 11 peaks at low 2θ needed for magnetic structure refinements). The sample was further cooled to 2 K below T_c and the same 131 reflections were measured again. In general, there was a clear increase in intensity on the low 2θ magnetic reflections at 2 K. Moreover, a temperature ramp from 2 K to 14 K around (200) reflection was measured as shown in figure 2. The measurement indicated that the strong magnetic peaks vanish at temperatures above 10.5 K, a result which is in good agreement with the magnetization measurement.

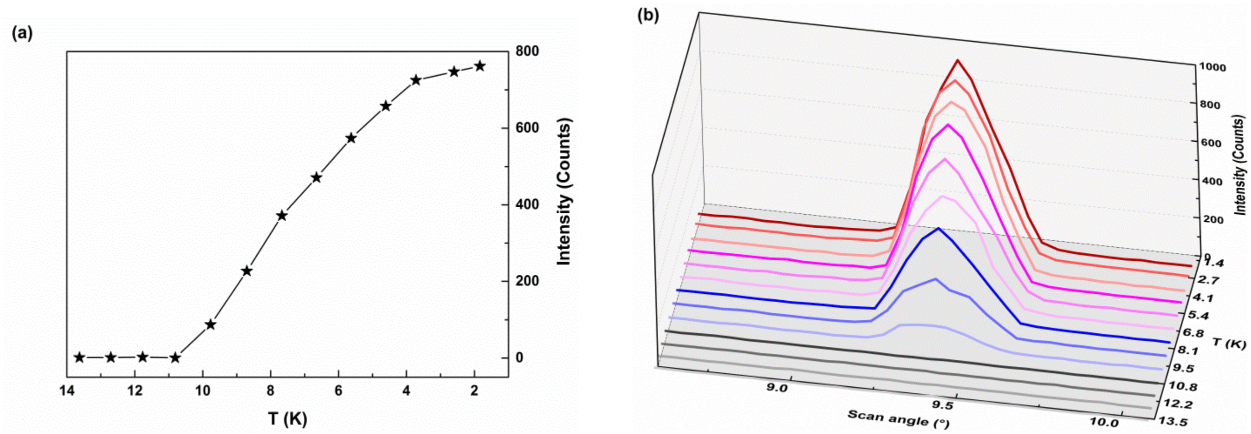

Figure 2: Plots of temperature scan vs diffraction intensity near T_c at the (002) reflection for a Tb-Au-Si AC: (a) shows integrated diffracted intensity vs temperature and (b) diffracted intensity profile in the scan angle range vs temperature.

The magnetic structure refinement of the Tb-Au-Si AC from the SCND data is in progress. However, in the preliminary results it is apparent that the magnetic moments of the Tb ions located at symmetrically equivalent positions in the nuclear structure gave different magnitudes as was previously reported from powder neutron diffraction refinement[8]. A careful analysis of the magnetic structure is underway, and it is clear that a larger number of reflections along with comparisons with isostructural compounds containing other Rare Earth elements (such as Ho-Au-Si) would be beneficial for a complete understanding of the magnetic behavior.

References

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