Experimental report

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Proposal: 5-41-879 **Council:** 4/2016

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

Research area: Materials

This proposal is a continuation of 5-41-842

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Samples: Au5Si1.3Ho

Instrument	Requested days	Allocated days	From	To
D9	5	7	03/10/2016	10/10/2016

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 and RE=Rare Earth), 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 5-41-879 Magnetic structure analysis of quasicrystal approximants in the Au-Si-Ho system, by single crystal neutron diffraction.

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Since the discovery of the first icosahedral quasicrystal (QC) by Shechtman et al. [1], their detailed atomic structures eluded researchers for many years until the recent structure solution of the binary icosahedral $Cd_{5.7}Yb$ quasicrystal [2]. The influence of long-range as well as short-range atomic order of icosahedral symmetry on the magnetic properties has in parallel been an exciting and fundamental issue in condensed matter physics and been investigated for almost thirty years [3-6]. As far as the magnetism of the rare-earth (*RE*) bearing QCs such as Zn-Mg-*RE* [3], Cd-Mg-*RE* [4], Cd-*RE* [7], etc., 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 [5]. However, it was recently shown that their crystalline counterparts, i.e., Cd_6*RE* approximant crystals (ACs), exhibit antiferromagnetic and ferrimagnetic transitions [8-11]. Recently we reported observations of ferromagnetic (FM) transitions in the Au-based compounds Au*SM-RE* (*SM*=Si,Ge) [12], which are ternary 1/1 cubic approximants related to the binary icosahedral $Cd_{5.7}Yb$ quasicrystal, and are composed of so-called Tsai-type clusters containing RE_{12} icosahedra of the magnetic elements [13,14]. The Cd_6*RE* approximants do not easily lend themselves to detailed investigations of magnetic structure because of the high absorption of Cd to neutrons. Since then, larger single crystals have successfully been grown of several Au-Si-RE (RE=Tb and Ho) approximants. Recently, we have performed single crystal neutron diffraction (SCND) measurements on an Au-Si-Tb approximant at the D10 beamline at ILL. The analysis of the data resulted in a model for the magnetic structure having unequal magnetic moments on Tb atoms which were symmetry equivalent in the nuclear structure. This clearly demanded further investigation and prompted us to study the related approximant in the Au-Si-Ho system where we had successfully produced large single crystals of very high quality (Figure 1).

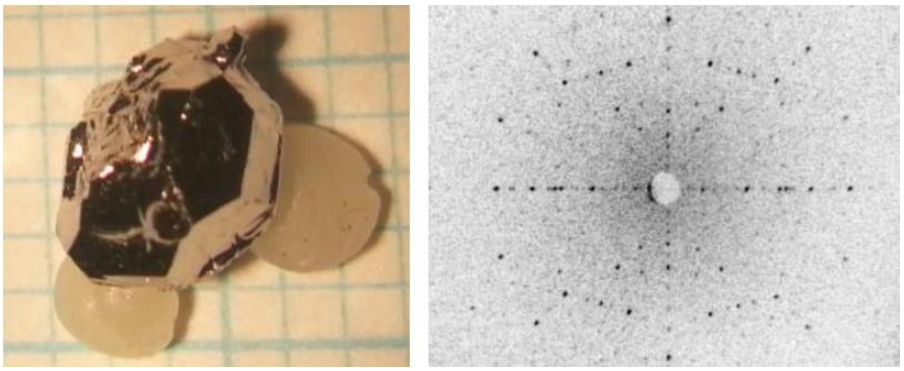

Figure 1: (Left) optical microscope image of a Au-Si-Ho single crystal; the scale is in mm. (Right) Laue neutron diffraction pattern along [1 0 0] for the Au-Si-Ho single crystal showing the excellent crystal quality for the proposed experiment.

The measurements on the Ho-containing sample were successful and a large number of nuclear and magnetic intensities could be collected in order to obtain good refinements of both the nuclear and magnetic structures. Two datasets were collected; one above the magnetic transition temperature and the other below. No additional magnetic peaks were detected but a clear intensity increase at the nuclear Bragg positions was detected below the transition temperature confirming the ferromagnetic nature of the sample with propagation vector k=0. Refinement of the magnetic structure was attempted on a dataset containing only the magnetic contributions to the Bragg intensities; which were extracted by subtracting the dataset collected above the magnetic transition temperature from that collected below. At first, refinements were done in every possible magnetic spacegroup related to the spacegroup Im-3 of the nuclear structure. The best refinement was obtained in the rhomohedral spacegroup R-3. Independently, we attempted to solve the magnetic structure by simulated annealing removing all symmetry. In the latter case a structure solution was obtained which clearly had a true symmetry that was higher than P1. The model was tested for higher symmetry, and the symmetry analysis revealed that the symmetry of the structure solution once again was R-3. In other words the same result was obtained by two different routes. This model gave a much better fit with the experimental data for both the Au-Si-Ho and Au-Si-Tb approximants than a previously proposed model obtained from powder data that assumed a simple collinear structure. Furthermore in this new model the problem of largely unequal moments on the RE-sites was solved and the magnetic moments agree well with what is expected for the corresponding RE-elements Tb and Ho. An image of the resulting magnetic structure is seen in Figure 2.

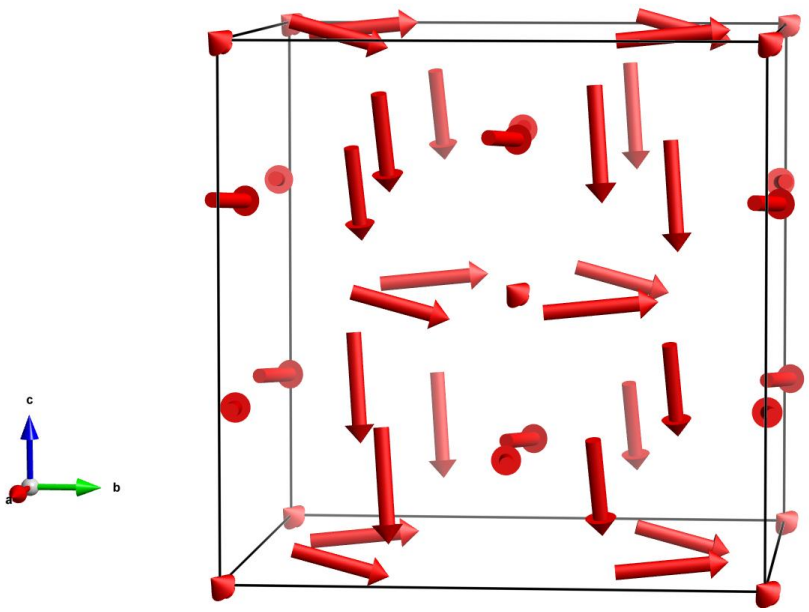

Figure 2: Magnetic structure of Au-Si-Ho. Only the magnetic moments of the Ho atoms are shown for clarity. All moments are equal except for the partially occupied RE-position at 0, 0, 0.