Experimental report

19/06/2019

Proposal: 5-41-938 **Council:** 4/2017

Title: Magnetic structure analysis of thequasicrystal approximant in the Au-Si-Gd system, by single crystal neutron diffraction.

Research area: Materials

This proposal is a continuation of 5-41-879

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Samples: Au5 Si1.3 Gd1

Instrument	Requested days	Allocated days	From	To
D9	12	12	22/06/2018	04/07/2018

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.

Magnetic and atomic structure solution of a Gd-Au-Si compound: a 6 X 2 X 2 superstructure of the basic Tsai-type 1/1 approximant structure.

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Abstract

Single crystal neutron diffraction experiment has been carried out on a Gd-Au-Si compound to determine its magnetic structure. Since ^{157}Gd is good neutron absorber, the experiment was done using short wave length neutrons (λ = 0.507 Å) at the D9 beam line at ILL. However, the atomic structure of the title compound is turn out to be a 6 X 2 X 2 superstructure of the basic Tsai-type 1/1 approximant crystal (AC) phase, for which the single crystal neutron diffraction experiment was initially intended. Hence, full magnetic structure determination was not be able to retrieve from the current neutron diffraction data which does not include superlattice reflections. The weak superlattice reflections could only be detected by single crystal x-rays diffraction experiment with a micro-focus source and longer exposure times. Attempts to measure the superlattice reflections during the neutron diffraction experiment did not succeed. Hence, the neutron diffraction data analysis considers only main reflections. Simulated annealing was employed on the neutron diffraction data collected below T_c (≈ 2 K) which has resulted to a solution different from the one previously obtained for the standard 1/1 ACs for a Ho-Au-Si system. In the current model, the magnetic moments have a tendency for collinearity which was not the case for the standard 1/1 AC. A temperature scan near the magnetic transition temperature (T_c) on a selected magnetic reflections clearly verify the difference in the magnetic behavior between the title superstructure and the standard ACs. The magnetic transition in the superstructure is very gradual while in the standard AC is sharp.

Results and discussions

Room temperature SCXRD experiment was performed on two single crystal specimen from the Gd-Au-Si sample. Preliminary results of the structure solution indicate that the compound is a 6 X 2 X 2 superstructure of the basic Tsai-type 1/1 AC. The super cell has a lattice parameter of 88.5 Å along the 6-doubled direction. We anticipate that the commensurate superlattice could be caused by an occupational wave affecting the Au/Si mixing ratio in the structure as clearly seen in the electron density map in figure 4.

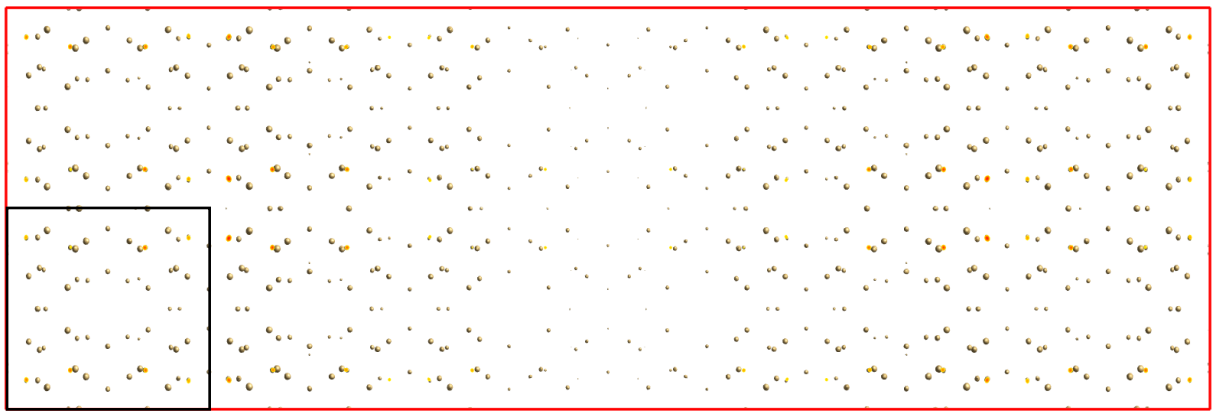

Figure 4: Electron density isosurfaces showing the density variation in the supercell (red) along the 88.5 Å period. The basic 14.72 Å cubic unit cell is outlined in black. In the most Si rich part of the structure the basic unit cell shrinks to a value of 14.56 Å as Au is replaced by the smaller Si.

The title compound shows magnetic long-range order at low temperatures. The magnetic property was first studied macroscopically by the SQUID magnetometer before the neutron experiment. Figure 5 shows magnetization as a function of temperature (M vs T) measurement results. The results indicate the presence of magnetic ordering at low temperatures below 25 K.

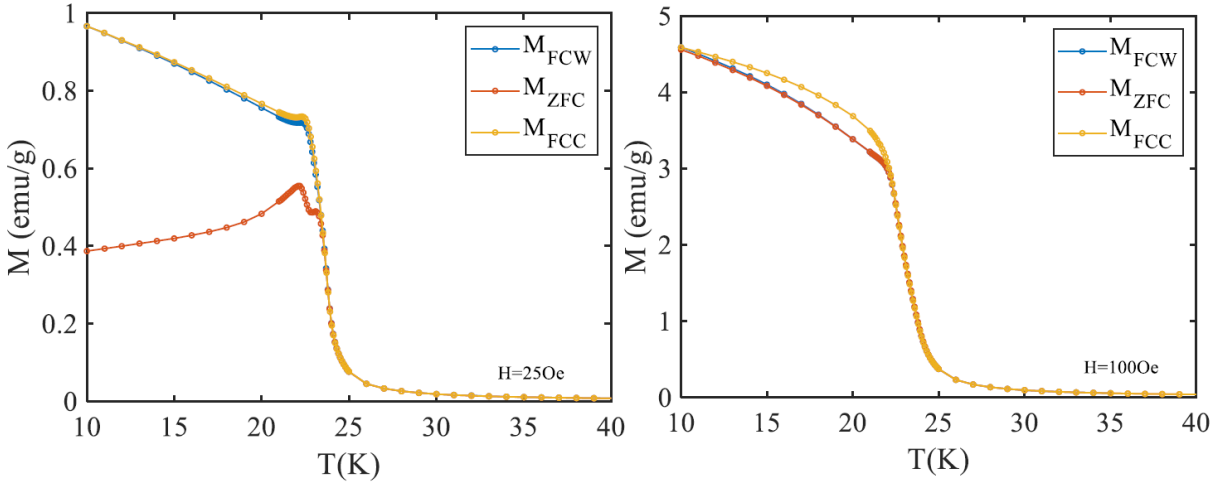

Figure 5: Magnetization (M) as a function of temperature (T) plots for the Gd-Au-Si superstructure with applied magnetic field of 250 Oe (Left) and 100 Oe (right). The notations in the insets of the figures: M_{ZFC}, M_{FCW} and M_{FCC}, refers to magnetizations measurements with zero field cooled, field cooled warm and field cooled cooled, respectively.

To confirm the macroscopic magnetic property result and to have a deeper insight especially on the microscopic magnetic behavior, SCND experiment was carried out on the Gd-Au-Si superstructure. The experiment was done first at 30 K, above the magnetic transition temperature (T_c) and later at 2 K below T_c at 2 K. The magnetic order is clearly observed below T_c as an increase of Bragg intensities of the main (Im-3) peaks. The magnetic transition behavior for the current compound is in contrast to the conventional basic Tsai-type 1/1 approximants previously studied by SCND experiments for example in the Ho-Au-Si and Tb-Au-Si systems. As shown in figure 5, the magnetic transition is not sharp in the title compound; it is seen as a gradual increase of the intensities over an extended region at low temperature. Similar measurements on the standard 1/1 approximants has shown sharp transitions.

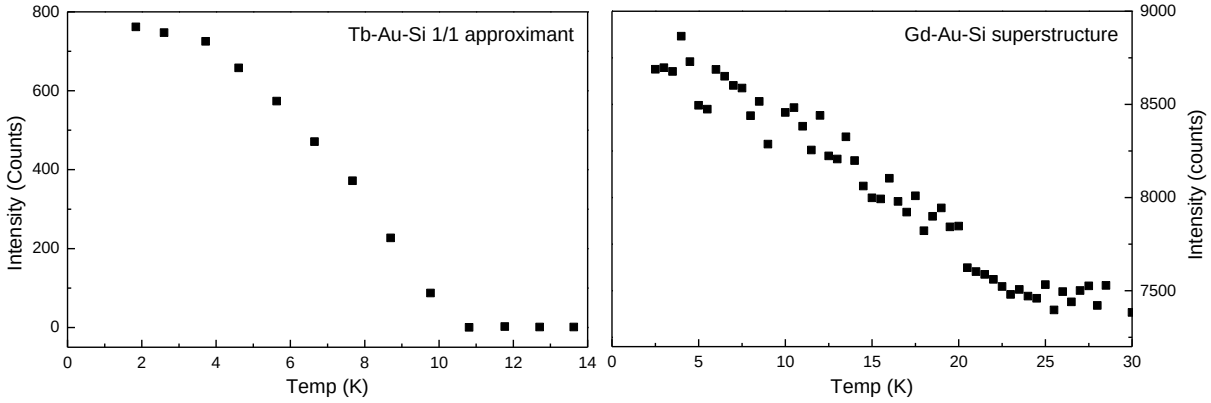

Figure 5: Comparison of single crystal neutron diffraction experiment results from a basic Tb-Au-Si approximant crystal and the Gd-Au-Si superstructure. Integrated intensity versus temperature scan from the (200) (left) and the (400) magnetic reflection (right). The transition is sharper in the basic 1/1 approximant than in the superstructure.

No measurable magnetic intensity contribution could be detected for any superlattice reflections, these were in fact not even detectable for the nuclear structure because of the high absorption of Gd and the low flux at 0.507 Å. Attempts to solve the basic magnetic structure in the spacegroup I-1 (non-standard, triclinic while keeping the centering translation at (½ ½ ½) give the following results: the magnetic structure solved by simulated annealing is different from the one observed for the Ho compound. The results however are yet inconclusive and better data is needed (either using a Gd isotope or reproducing the synthesis using a different RE metal). The amplitudes of the obtained moments are close to 7.7 μ_B which is in good agreement with the theoretical value for Gd^{3+} (7.94 μ_B).

The magnetic structure obtained from SA alongside the refined one for the standard Tsai-type 1/1 Ho-Au-Si AC are shown below for comparison.

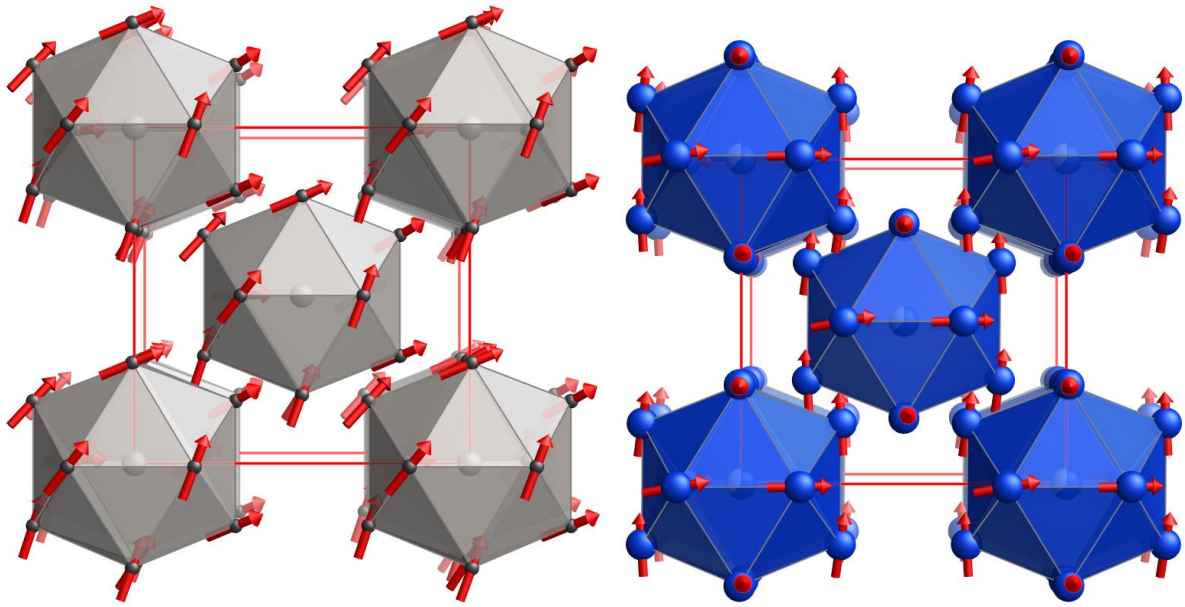

Figure 6: Magnetic structure solution obtained from simulated annealing of SCND data for the Gd-Au-Si superstructure (left) and basic 1/1 AC of the Ho-Ai-Si system (right). The magnetic structure of the Gd-Au-Si compound has a stronger tendency towards collinearity.