Experimental Report

29/01/2014

Proposal: **7-01-371** **Council:** 10/2012

Title: Tetrahedron dynamics in a series of approximant crystals to the icosahedral quasicrystal i-AlCuSc

This proposal is resubmission of: 7-01-354

Research Area: Physics

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Samples: MgScZn
Al40.5Cu45.0Sc14, Al36.4Cu48.1Sc15.5
Cd6T6

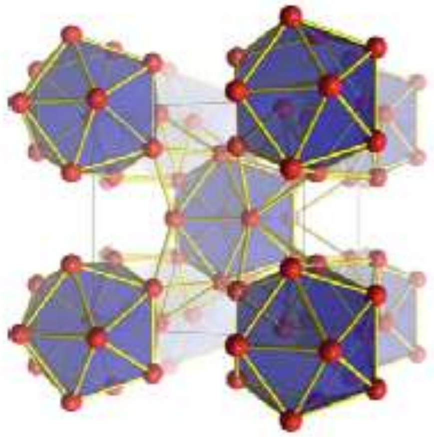
Instrument	Req. Days	All. Days	From	To
IN6	0	6	02/04/2013	08/04/2013
IN4	5	5	27/05/2013	01/06/2013

Abstract:
The AlCuSc system features approximant phases to icosahedral quasicrystals. These phases have almost identical composition and consist of the same cluster building blocks (Tsai type clusters). The innermost shell of these clusters is a symmetry breaking tetrahedron which has been shown to reorient dynamically. The approximant phases differ in the exact arrangement of the Tsai type clusters, thus giving rise to different reorientation schemes of the tetrahedral shells. Therefore it becomes possible to investigate the direct influence of the cluster arrangement on the tetrahedron dynamics as well as the changes in the tetrahedron-tetrahedron correlations-mechanisms that come into play for stabilizing quasicrystals and approximants

Experimental report: IN6/IN4 experiment 7-01-371-132

Measurements of the magnetic structure and crystal field excitations of the quasicrystal approximant Cd_6Tb

Introduction



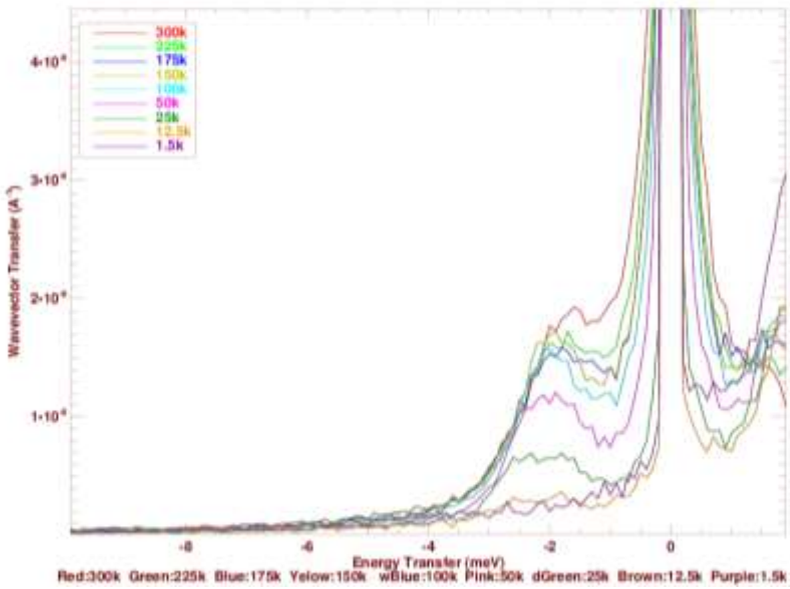
The possibilities for magnetic ordering in aperiodic structures have been the subject of intense theoretical and experimental studies since the discovery of icosahedral quasicrystals (QC) in 1982 [1]. However, all the known quasicrystals with local moments exhibit frustration and spin glass-like behavior at low temperature [2-3]. The origin of this frustration is strongly debated.For instance, the magnetic rare earth sites in Cd-Mg-R correspond to the vertices of the triangular faces of the icosahedral shells within Tsai-type clusters, forming the backbone of the quasicrystalline structure [4-5]. Here the geometrical frustration associated with triangular arrangements of spins follows naturally. The absence of long-range magnetic order is also evidenced in many periodic approximants of icosahedral QCs (approximants are periodic crystals with unit cell atomic decorations that are closely related to the respective QC phases). However, the 1/1 approximant to the Cd-Mg-R icosahedral phases, Cd_6Tb appears to be an exception to these findings. At ambient temperature, the rare earth ions of Cd_6Tb can be viewed in terms of a bcc packing of icosahedral clusters (see **Fig. 1**). Below 160 K, there is a small distortion from cubic to monoclinic such that the symmetry is reduced to C2/c. Nevertheless, the essential elements of the magnetic structure may still be discussed in terms of the cubic high temperature (HT) phase. At low temperature the total magnetic moments of the icosahedral shells have been shown to orient in such a way that anti-ferromagnetic order is achieved (T_N~25K) [6-7].

Experimental settings IN6

We have carried out an Inelastic Neutron Scattering (INS) experiment at the IN6 Time of Flight (ToF) spectrometer, investigating a ^{112}Cd isotope enriched sample of Cd_6Tb. We have measured elastic and quasielastic neutron scattering signal around strong Bragg reflections using a wavelength of 5.12 Å. The measurements have been performed on the Cd_6Tb 1/1 approximant to investigate (a) the nature of the magnetic ordering at low temperature and the local environment of the rare earth site via a measurement of the crystal field excitations (b) the dynamics (jumping) of the tetrahedra located at the center of the Tsai type cluster.

Experimental Results IN6

The powder averaged dynamical structure factor, which was measured with high energy resolution at the IN6 spectrometer, yielded a clear signature of crystal field excitations. This is clearly visible when data measured between 1.5 K and 300 K are compared to each other (see **Fig 2**). The temperature dependence of the evidenced excitations at 2-3 meV is consistent with what is expected for a crystal field splitting: the intensity of the excitation increases rapidly above 12 K and then saturates at about 100 K. The width of the 2 meV excitation is T independent. The position of the excitation is slightly changing with T and seems to get shifted towards lower energies when the structural phase transition sets in. At the structural phase transition a quasielastic signal related to the tetrahedron reorientation is expected. Yet, despite the fact that small changes in the quasielastic signal are evidenced between 150 K (below Tc) and 175 K (above Tc) this cannot clearly be attributed to the tetrahedron motion since the quasielastic spectrum is dominated by a magnetic contribution which sets in above 25 K thus being related to the AFM phase transition (see **Fig 2**)..



<u>Figure 2:</u> Q-Integrated S(Q,E) from IN6 for differences temperatures, sample Cd_6Tb

Experimental settings IN4

As follow up experiment we have carried out an Inelastic Neutron Scattering (INS) study on the IN4 Time of Flight (ToF) spectrometer, investigating a ^{112}Cd isotope enriched sample of Cd_6Tb.. We have measured elastic and quasielastic neutron scattering signal around strong Bragg reflections over a wide energy range, using different wavelengths (1.5 Å, 3 Å, 3.4 Å and 3.6 Å) to obtain complementary data to the recent IN6 experiment. Measurements have been performed on the Cd_6Tb 1/1 approximant to investigate the magnetic ordering and the crystal field splitting on the Stokes and the Anti-Stokes site. Moreover we aimed to extract a quasielastic signal corresponding to the dynamics (jumping) of the tetrahedra located at the center of the Tsai type clusters. We have measured the S(Q,E) for a temperature range from 1.8k to 300K with an orange cryostat while a cryoloop was used for the measures above to 400K.

Results and Discussion IN4

The experimental data again evidenced a clear magnetic quasielastic signal above 25 K while below 25 K no such signal is present thus pointing to the elsewhere evidenced antiferromagnetic order below 25 K (see **Fig. 2**) and thus corroborating the findings of the IN6 experiment. Moreover the measured S(Q,E) signal revealed a clear signature of the crystal field excitations, resulting in a double peak between 2 and 3 meV (see **Fig. 2**). Especially at low temperatures it could clearly be evidenced that the crystal field excitation has to components. Moreover, this double peak is again found to shift slightly to lower energies when the transition temperature of the structural phase transition at about ~160 K is reached. The tetrahedron jumps which were expected to be present above 160 K are masked by the magnetic quasielastic signal and also for high Q-values a clear distinction of quasielastic and the magnetic quasielastic signal have not yet been possible.

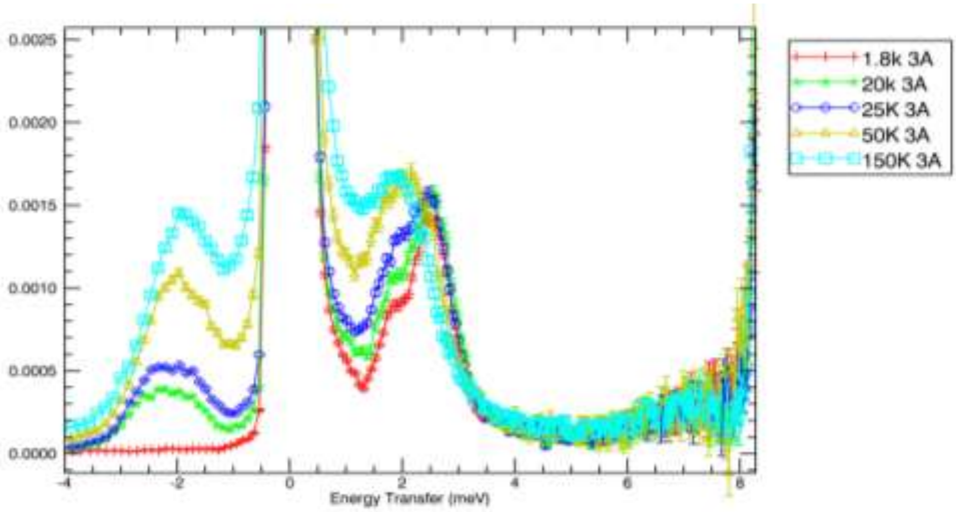


Figure 2 : **S(E) integrated on Q range at 3 Å, sample Cd_6Tb**

Summary

The measurements at IN4 and IN6 clearly evidenced a magnetic quasielastic signal above 25 K, while the excitations at 2-3 meV could clearly be attributed to the occurrence of crystal field splitting. The exact temperature dependence of the crystal field excitation energy will help to determinine the CEF anisotropy and possibly the direction of magnetic moments in the ordered antiferromagnetic state at lower temperatures. The tetrahedron jumps which were expected to be present above 160 K are, even for the higher Q-values accessible at IN4, masked by the magnetic quasielastic signal and thus a clear distinction of quasielastic and the magnetic quasielastic signal have not yet been possible.

References

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