実験報告書様式(一般利用課題・成果公開利用)

MLF Experimental Report	提出日 Date of Report 2017/7/26
課題番号 Project No. 2017A0233 実験課題名 Title of experiment Crystalline electric field excitation of icosahedral quasicrystal and its approximants, Au–Si–Pr and Au–Ga–Tb: Role of 5-fold symmetry and quasiperiodicity on magnetism 実験責任者名 Name of principal investigator Takanobu Hiroto 所属 Affiliation The University of Tokyo	装置責任者 Name of responsible person Shinichi Itoh 装置名 Name of Instrument/(BL No.) HRC (BL12) 実施日 Date of Experiment 2017/05/27 – 2017/5/31

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。(適宜、図表添付のこと)
Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

We measured three samples of Au-Si-Pr and Au-Ga-Tb approximant crystal and Au-Ga-Tb quasicrystal

Au-Si-Pr approximant; Au69 Si 17 Pr 14

Au-Ga-Tb quasicrystal; Au65 Si 20 Tb15

Au-Ga-Tb approximant; Au65 Si 21 Tb14

Approximants samples were prepared by arc-melting and subsequent annealing method, whereas quasicrystal sample was prepared by melt spinning method. All the samples were crushed into the powder (less than 100 μm).

2. 実験方法及び結果 (実験がうまくいかなかった場合、その理由を記述してください。)
Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

We measured the crystalline electric field excitation on the three intermetallic compounds, i.e., Au-Si-Pr approximant crystal (AP), Au-Ga-Tb quasicrystal (QC) and Au-Ga-Tb AP, by HRC beamline (BL12). The previous measurements on the Au-Si-Tb AP, that is isostructural to the Au-Si-Pr, we found the evidence for the unique crystal electric field (CEF) splitting effect originate from pseudo 5-fold symmetry around rare-earth ions. In order to future investigation, we focus on the two-main important point;

I. Comparison of 5-fold symmetry CEF by neglecting 5-fold effect (Pr^{3+} ion with J = 4 vs previously measured Tb^{3+} ion with J = 9)

II. Comparison of 5-fold symmetry CEF between periodic system and quasiperiodic system.

Approximant samples were prepared by conventional arc-melting method from raw elements following heat-treatment method, whereas quasicrystal sample were prepared by melt-spinning technique, due to its thermal instability. Phase purity was checked by means of powder XRD method. The magnetic susceptibilities were measured by a superconducting quantum interference device (SQUID) magnetometer (Quantum Design MPMS-XL); Au-Ga-Tb QC and Au-Ga-Tb AP shows a magnetic transition or anomaly at 12.5 K (QC) and 13.1

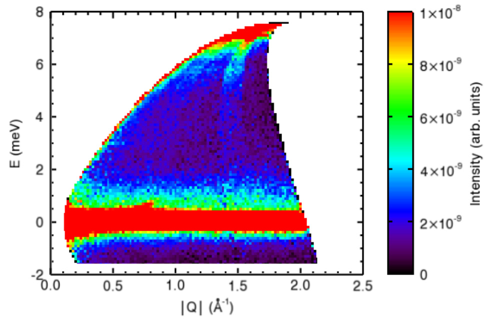
2. 実験方法及び結果（つづき） Experimental method and results (continued)

K (AP), receptibility. On the other hand, no magnetic transitions ware observed for Au-Si-Pr AP down to 2K.

Neutron scattering experiments were performed by HRC beamline (BL12). Each powdered sample (~ 12 grams) were installed inside double annual aluminum cell in order to earn the effective volume. Temperature range for measurements were between 3 K and 100 K.

<u>I. Comparison of 5-fold symmetry CEF by neglecting 5-fold effect</u>

Previous Au-Si-Tb AP shows a broad CEF peak centered around 4.5 meV. On the other hand, for Au-Si-Pr AP shows no clear CEF beak was observed. (Figure 1) At higher temperature, we observed that only quasielastic signal widens. Furthermore, we also found the inelastic signal at high energy mode around 180 meV with Ei = 385meV (Figure 2), that may be explained by the intermultiplet transition of Pr^{3+}. However, its Q-dependence is seemingly different from that observed in the metallic Pr.



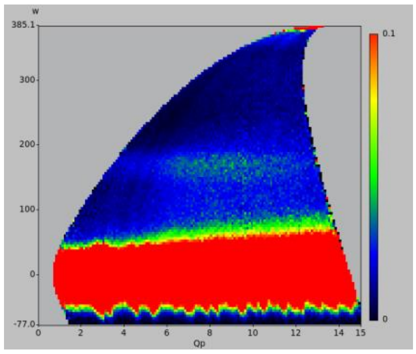
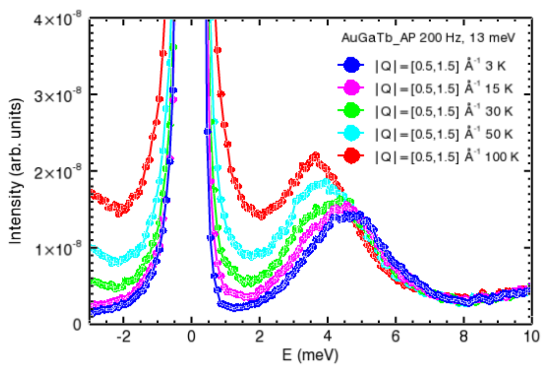


Figure 1(Left): S(Q,E) plot for Au-Si-Pr AP measured at Ei = 7.6 meV measured at 3 K.

Figure 2 (Right): S(Q,E) plot for Au-Si-Pr AP measured at Ei = 385 meV measured at 3 K.

<u>II. Comparison of 5-fold symmetry CEF between periodic system and quasiperiodic system.</u>

As expected, we found clear CEF peak centered around at 4.5 meV (for AP, figure 3) and 4 meV (for QC, figure 4). Its temperature dependence is very similar to the previous measurement for Au-Si-Tb approximant case. However, the width of CEF peak in quite different between QC and AP samples. This broaden CEF signal may be understood the distribution difference of Tb site (Tb atoms occupy only one crystallographic site for AP sample, whereas Tb atoms occupy several sites for QC case). If this picture is true, our observations is the first demonstration of the ‘quasiperiodicity’ through the magnetism.



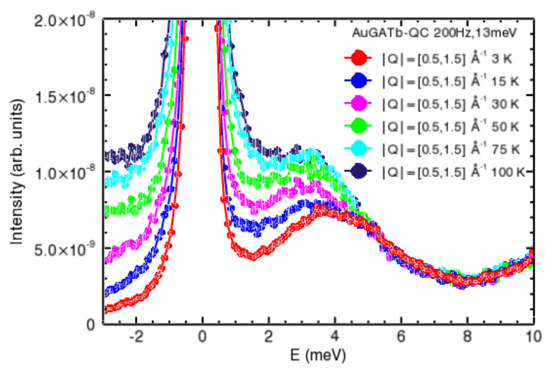


Figure 3(Left, approximant) and 4(Right, quasicrystal): Temperature dependence of inelastic signal for Au-Ga-Tb AP and QC measured at Ei=13 meV.