Growth and characterization of Al-Cu-Ru icosahedral quasicrystals from self-fluxes

Kotoba Toyonaga[1], Tsunetomo Yamada[2], Marc de Boissieu[3], Olivier Peréz[4], Pierre Fertey[5] and Hiroyuki Takakura[6]

[1]Graduate School of Engineering, Hokkaido University, Sapporo, Hokkaido, 060-8628, Japan

[2]Faculty of Science, Department of Applied Physics, Tokyo University of Science, 6-3-1 Niijuku, Katsushika-ku, Tokyo, 125-8585, Japan

[3]Universite Grenoble Alpes, SIMaP, CNRS, Gerenoble INP, Grenoble F-38000, France

[4] CRISMAT, CNRS UMR 6508, 6 Bd Marechal Juin, F-14050 Caen Cedex 4, France

[5] Synchrotron SOLEIL, Gif-sur-Yvette F-91192, France

[6]Division of Applied Physics, Faculty of Engineering, Hokkaido University, Sapporo, Hokkaido, 060-8628, Japan

s26153022d_k_toyo@eis.hokudai.ac.jp

Several atomic structure models for F-type Al based icosahedral quasicrystals (iQCs), such as Al-Cu-Fe and Al-Pd-Mn iQCs have been reported so far [1,2]. However, these iQCs have qualitative problems, for example, containing linear phason strains and/or producing lots of diffuse scatterings, and insufficient number of Bragg reflections have been observed for the analysis of their atomic structures. Therefore, it can be said that the atomic structure model of this category of iQCs has not been established yet. In this study, we focus on the Al-Cu-Ru iQC to investigate the atomic structure of F-type Al based iQCs. Obtaining a good quality iQC in this alloy system is considered likely, because of the absence of linear phason strains [3]. Although a growth of single QC has been reported by using self-flux method [4], it is not clear whether there is a difference in quality depending on the growth conditions. We have investigated the conditions for obtaining a good quality Al-Cu-Ru iQC by using the self-flux method.

Several master alloys having the same composition, $Al_{57.0+x}Cu_{39.5-x}Ru_{3.5}$ (x = 0, 5, 10) or $Al_{62.0}Cu_{34.0+y}Ru_{4.0-y}$ (y = 0, 0.5, 1.5), were prepared by arc melting. Each alloy was inserted in an alumina crucible and sealed in a quartz tube with high purity Ar. The tubes were heated up to 1150℃ and kept for 2 h, then, subsequently cooled to various temperatures in a range of 800-1000℃ with the cooling rate of 2 K/h. When they were reached at the targeted temperatures, they were quenched in water or held at the temperatures for various durations in a range of 5-750 h. With this protocol, a bunch of single iQCs having several mm in size has been obtained respectively. The iQCs obtained were studied by powder X-ray diffraction (XRD), electron diffraction, Electron Probe Micro Analyzer (EPMA) and single crystal XRD. The Bragg intensity data by single crystal XRD were collected at CRISTAL beamline at synchrotron SOLEIL. The structure solution was obtained by Superflip [5] and the six-dimensional electron densities were calculated by using qcmem [6].

We summarize here the results obtained for the master alloys of $Al_{62.0}Cu_{34.5}Ru_{3.5}$. The compositions of iQCs obtained with the conditions of 800℃-300 h, 900℃-750 h and 1000℃-5 h were $Al_{64}Cu_{21}Ru_{15}$ (a_{ico}=9.049(5)Å), $Al_{65}Cu_{20}Ru_{15}$ (a_{ico}=9.048(1)Å) and $Al_{67}Cu_{16}Ru_{17}$ (a_{ico}=9.072(1)Å), respectively. The iQCs obtained by the condition of 800℃ showed slightly wider peak widths than those obtained at higher temperatures, and no improvement had been observed with varying the holding time. On the other hand, iQCs obtained at higher temperatures (900℃ and 1000℃) showed narrower, approximately 20-60% smaller, peak widths. Fig. 1 compares the peak widths of powder XRD from iQCs grown with the conditions of 800℃-300 h and 1000℃-5 h, and those reported previously [3]. We conclude that Al-Cu-Ru iQCs with best quality were obtained by the condition of 1000℃-5 h in the present study. No linear phason strains were observed for all iQCs studied by powder XRD.

Single crystal XRD experiments (λ=0.58337Å) have been conducted for a piece of single grain iQC grown by the condition of 1000℃-5 h. Around 2700 independent Bragg reflections ($I > 3\sigma(I)$) were obtained. Subsequently, we have tried a phase retrieval of the diffraction data. The structure solution has been obtained successfully with the value of R = 14.593%. Fig. 2 shows the corresponding electron densities, which clearly indicate a characteristic of F-type structure and periodic arrangement of occupation domains in six-dimensional space.

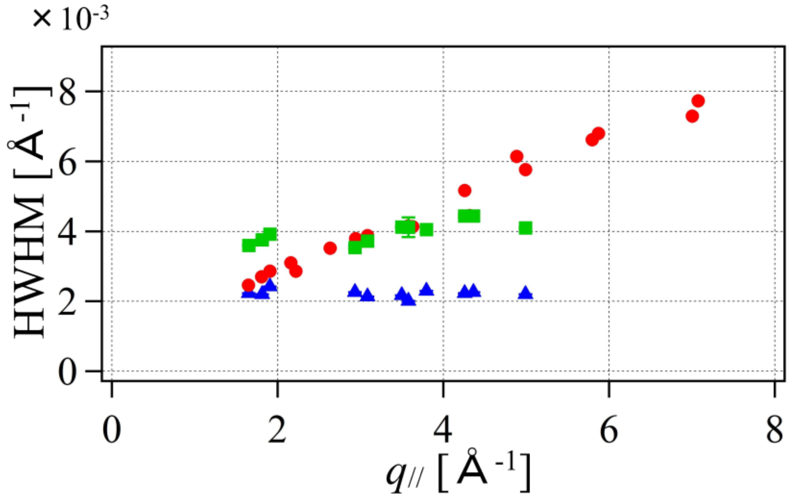

Figure 1. The HWHMs of powder XRD from Al-Cu-Ru iQCs grown with the conditions of 800℃-300 h (green squares) and 1000℃-5 h (blue triangles) in the present study and those reported previously (red circles) [3].

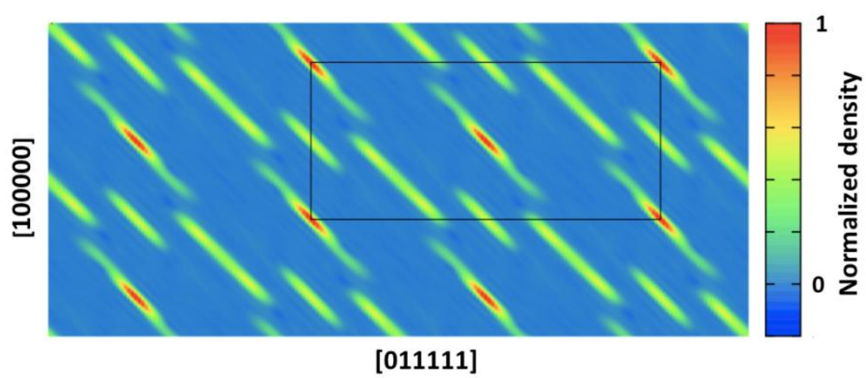

Figure 2. The two-dimensional section of the six-dimensional electron densities cut by plane containing fivefold axes both in the external and internal spaces for $Al_{67}Cu_{16}Ru_{17}$ iQC grown with the condition of 1000℃-5 h. The black solid line indicates the section of unit cell in six-dimensional space.

1. A. Katz & D. Gratias, *J. Non-Cryst. Solids*, **153**&**154**, (1993), 187

2. A. Yamamoto *et. al.*, *Phys. Rev*. B, **68**, (2003), 094201.

3. C. A. Guryan *et. al.*, *Phys. Rev. Lett.* B, 62, **20**, (1989), 2409.

4. J. Q. Guo *et. al.*, *J. Crystal Growth*, **236**, (2002), 477.

5. L. Palatinus & G. Chapuis, *J. Appl. Cryst.* **40**, (2007), 786.

6. A. Yamamoto, *Sci. Technol. Adv. Mater*. **9**, (2008), 013001.