Now showing in 4D: Quasicrystal growth and dissolution

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How does a quasicrystal grow? Although there has been no lack of theoretical studies on quasicrystal growth, there have been very few experimental investigations with which to test their various hypotheses. To this end, we have recently performed synchrotron-based experiments on a decagonal (*i.e.*, two-dimensional or 2D) quasicrystal of composition Al-8wt%Co-8wt%Ni [1]. High flux X-ray tomography enabled us to capture projections at temperature during continuous cooling. By reconstructing the data in three dimensions (3D) as a function of time (i.e., a 4D measurement), we were able to obtain new insights on the growth and dissolution pathways of the quasicrystals and related approximant phases.

During growth, the ten-fold symmetry of the 2D decagonal quasicrystal is self-similar, indicating that the bounding aperiodic facets propagate at roughly the same rate (see **Fig. 1(a)**). In this mobility-limited regime, growth likely occurs *via* cluster attachments and rearrangements at the growth front. However, the ten-fold symmetry is broken upon dissolution (**Fig. 1(b)**). *Therefore, the growth and dissolution processes of the decagonal quasicrystal lack time-reversal symmetry*, and quasicrystal dissolution is not locally-controlled [1]. Instead, dissolution is limited by buoyancy-driven convection in the surrounding melt, leading to highly anisotropic growth rates of the bounding aperiodic facets.

In addition, we have calculated the kinetic coefficient — defined as the constant of proportionality between interface velocity and undercooling — of the Al-Co-Ni decagonal quasicrystal during the growth process. The calculated kinetic coefficient of the Al-Co-Ni decagonal quasicrystal is significantly smaller than that of periodic crystals (both simple and intermetallic) by over three orders-of-magnitude (**Fig. 1(c)**) [1]. The smaller kinetic coefficient points to the presence of large attaching clusters or "building blocks" that contribute to a more sluggish growth rate. This observation supports the theory of cluster-based quasicrystal growth. *Broadly, the kinetic coefficient is a signature of the structural complexity of a crystal.*

Comparisons of the growth dynamics are also drawn between decagonal, icosahedral [2], and approximant [3] phases. The kinetic coefficient of an approximant phase in the Al-Co-Ni system is even smaller than that of a quasicrystal, indicating that the latter is structurally more flexible than the former. Altogether, by extracting morphological, dynamic, and compositional information directly from our space- and time-resolved data, we were able to provide a fresh lens on these poorly understood phase transitions. Our *in situ* studies provide the much-needed benchmark data that can be used to validate simulations and theories of the dynamic behaviors of complex intermetallics, including quasicrystals.

1. I. Han, X. Xiao, and A.J. Shahani, *Sci. Rep.*, **7**, (2017), 17407.

2. N. Senabulya, I. Han, X. Xiao, and A.J. Shahani, *Scripta Mater.*, **146**, (2018), 218.

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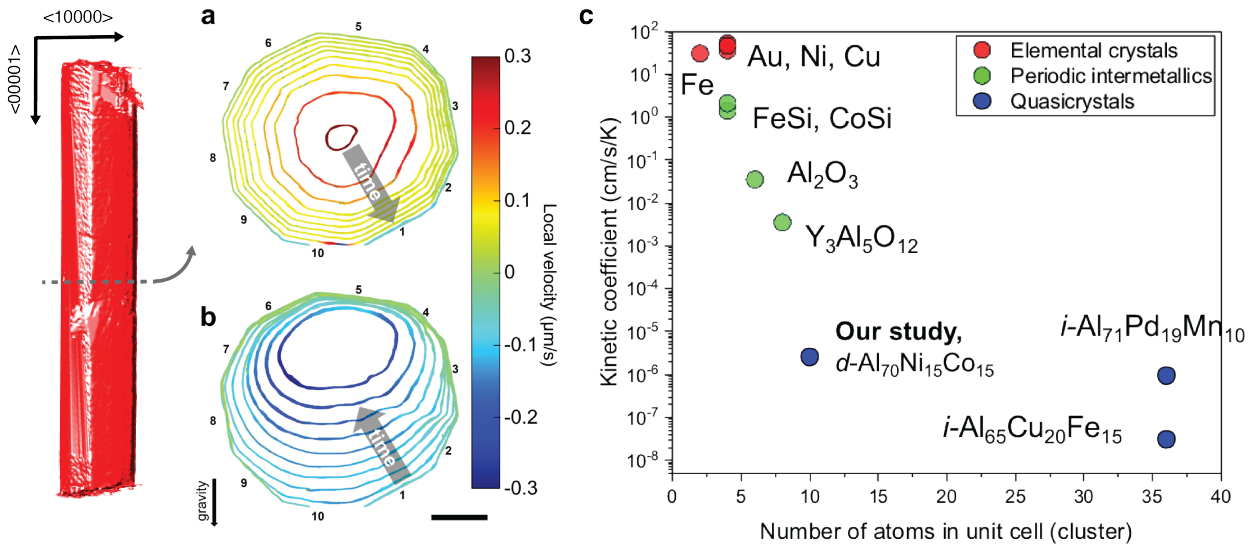

Figure 1. 4D microstructural evolution of a decagonal quasicrystal of composition Al-8wt%Co-8wt%Ni. The structure consists of a periodic <00001> "long axis" and perpendicular, aperiodic <10000> directions. Interfacial isochrones of the solid-melt interface in the ten-fold {00001} plane during (a) growth and (b) dissolution, colored according to the interface velocity. Positive velocity corresponds to growth and negative to dissolution. (c) Growth kinetic coefficients of various crystals (elemental, periodic intermetallic, and quasicrystalline) *vs.* number of atoms in unit cell or cluster. Quasicrystals of both decagonal *d* and icosahedral *i* variants possess kinetic coefficients that are at least three orders-of-magnitude smaller than that of other known periodic crystals. Adapted from Ref. [1].