

## Supplemental Movie Captions

1. Segregated homogeneously nucleated PP' crystal. The green is the major P species, while the red corresponds to the impurity, P'. Note that the few red particles in the crystal are segregated to the surface. This video corresponds to Figure 4a.
2. Well incorporated homogeneously nucleated PP' crystal. The green in the major P species, while the red corresponds to the impurity, P'. Note that the red particles are dispersed throughout the crystal – though a non-negligible fraction is segregated to the surface. This video corresponds to Figure 4b.
3. Well incorporated PP' crystal formed *via* sedimentation. The green in the major P species, while the red corresponds to the impurity, P'. Note that the red particles are well dispersed throughout the exceptionally large crystal. This system is ideal, as it boasts well incorporated impurities as well as large crystalline domains, which mean fewer defects. This video corresponds to Figure 4c.
4. Well incorporated homogeneously nucleated ABB' crystal. The green in the major A species, while the red corresponds to the impurity, B'. B particles are not dyed. The red particles are dispersed throughout the system, due to an adequate (<.5 degree) temperature match between A-B and A-B'. This video corresponds to Figure 4e.
5. Z-stack video of a PP' Icosahedral cluster resting on a triangular face. To the left is a scan of the cluster, with green corresponding to P particles, and red corresponding to P'. The middle image is a rendering of the cluster based on manual centroiding. The right-most image is a wire-frame projection of the cluster. This video corresponds to Figure 5a.
6. Z-stack video of an AB' Icosahedral cluster resting on a two-particle edge. To the left is a scan of the cluster, with green corresponding to A particles, and red corresponding to B'. The middle image is a rendering of the cluster based on manual centroiding. The right-most image is a wire-frame projection of the cluster. This video corresponds to Figure 6a.
7. Z-stack video of an AB' n+10 Tammes cluster resting on a square face. To the left is a scan of the cluster, with green corresponding to A particles, and red corresponding to B'. The middle image is a rendering of the cluster based on manual centroiding. The right-most image is a wire-frame projection of the cluster. This video corresponds to Figure 6d.
8. Z-stack video of an AB' cubical cluster resting on a vertex. To the left is a scan of the cluster, with green corresponding to A particles, and red corresponding to B'. The middle image is a rendering of the cluster based on manual centroiding. The right-most image is a wire-frame projection of the cluster. This video corresponds to Figure 7b.
9. Z-stack video of an AB' skew-cube cluster resting on a square face. To the left is a scan of the cluster, with green corresponding to A particles, and red corresponding to B'. The middle image is a rendering of the cluster based on manual centroiding. The right-most image is a wire-frame projection of the cluster. This video corresponds to Figure 7c.