Size-control and escape mechanisms in self-limiting assemblies with open boundaries

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Brandeis bioinspired MRSEC

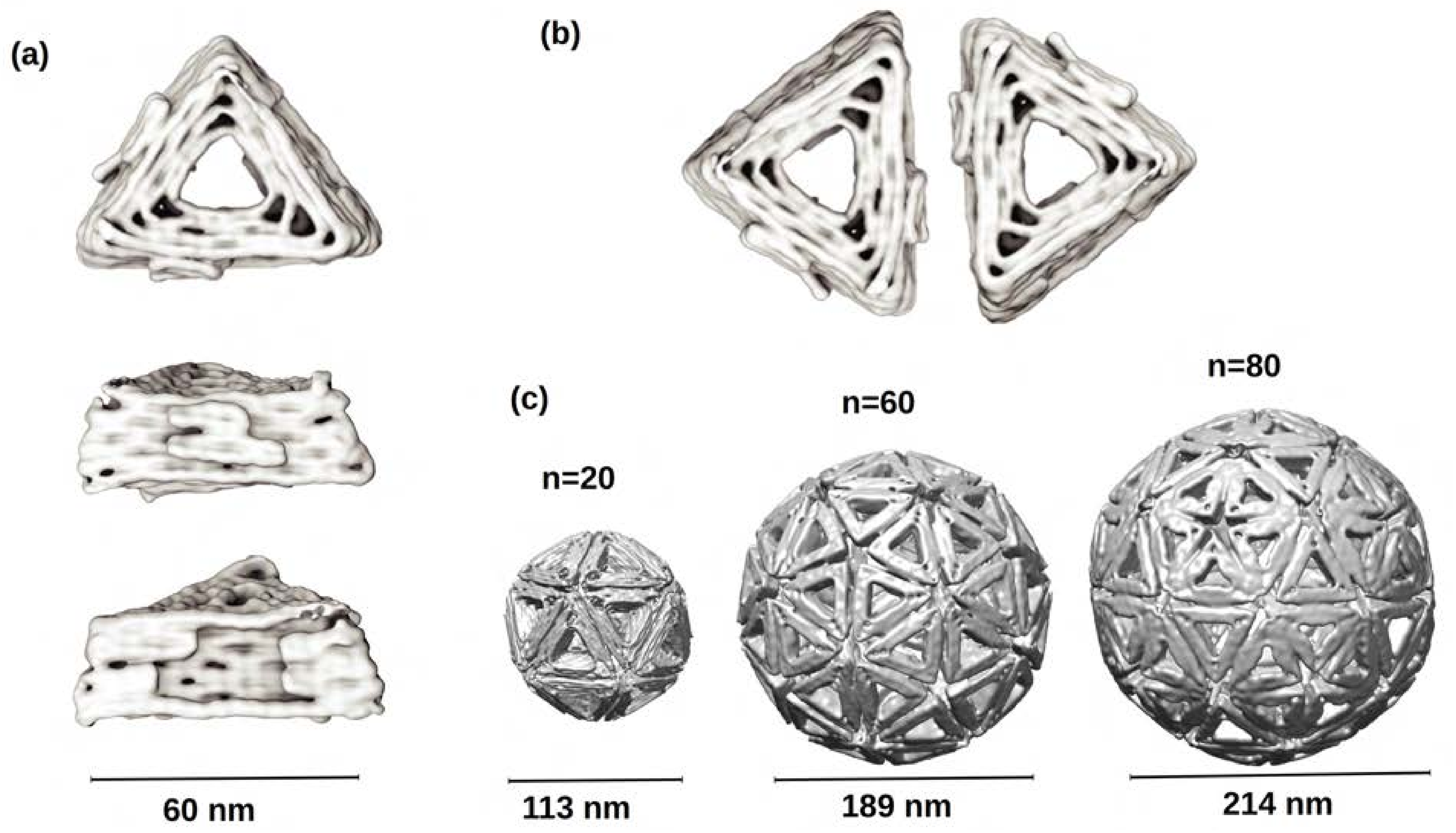
Abstract

The self-limited assembly (SLA) of building blocks into structures with large, but well-defined finite sizes, is an essential capability of biological systems. Developing human-engineered building blocks with a similar ability to undergo SLA is of great interest in nanotechnology and is one of the main objectives of MRSEC's IRG1. Recently, it has been theoretically proposed that SLA can be achieved through geometric frustration, in which the preferred local packing of subunits is incompatible with their preferred large-scale assembly structure.

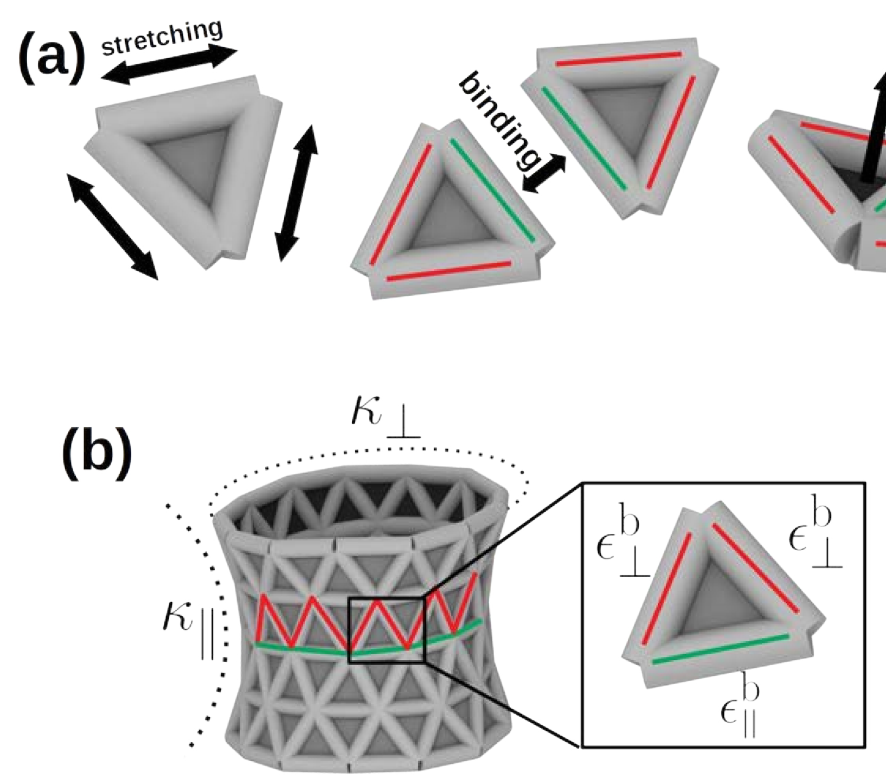
In this work we describe simulations of geometrically frustrated assembly motivated by ongoing experiments on DNA origami building blocks, carried out within IRG1. We consider triangular subunits that self-assemble into a hexagonal array, but have interaction geometries that favor formation of catenoid structures. The resulting negative Gaussian curvature is incompatible with hexagonal order, leading to geometric frustration. We use dynamic Monte Carlo simulations and free energy calculations to demonstrate that this incompatibility leads to equilibrium SLA in some parameter regimes, and identify two mechanisms by which the system escapes self-limitation in other regimes. The objective of the analysis is to support further DNA origami experiments; for example, by identifying the most important experimental control parameters and identifying conditions under which SLA is experimentally achievable in these systems.

Introduction

Self limited via self closing



Model for geometrically frustrated self assembly



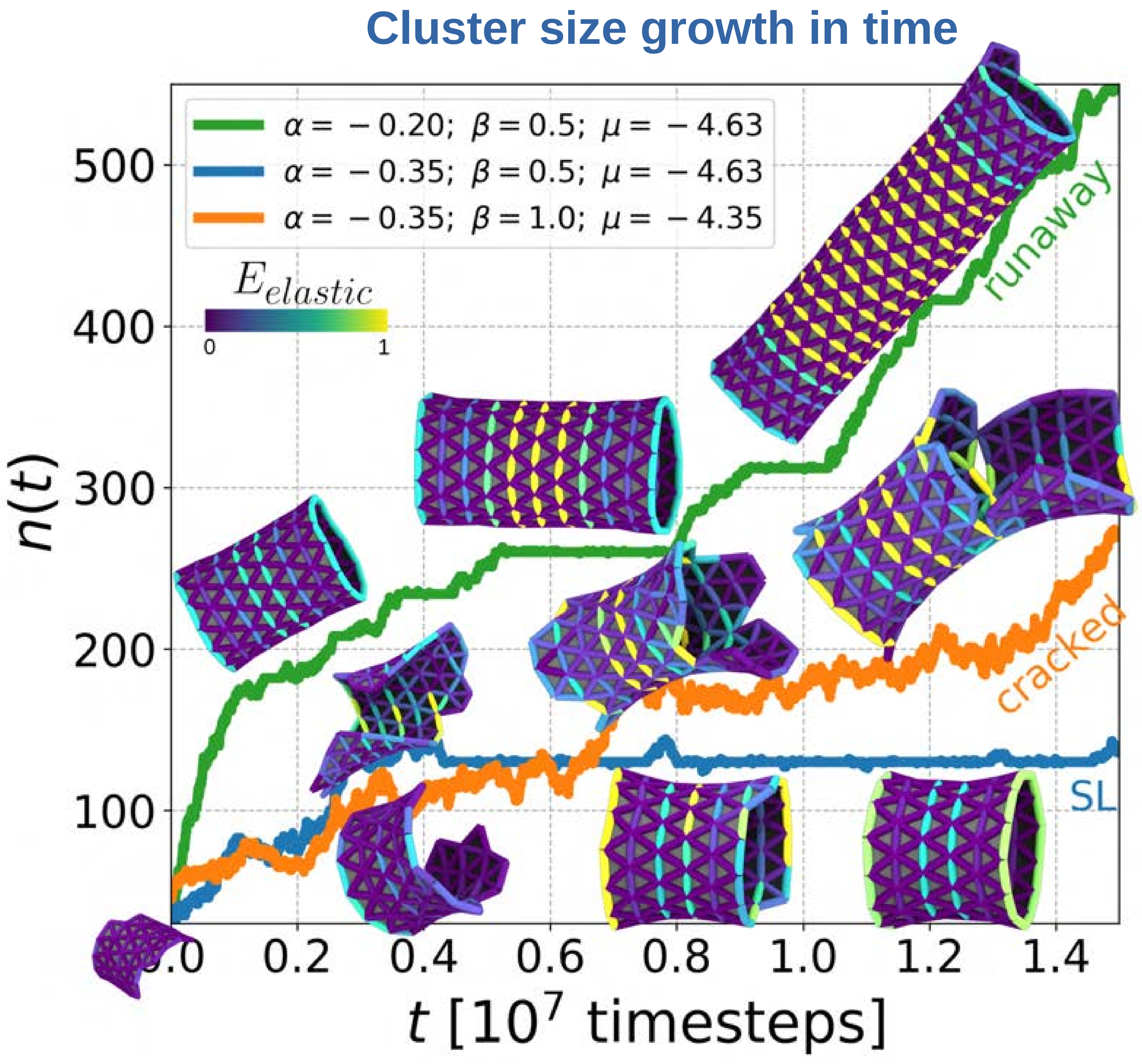
Curvature anisotropy: $\alpha = \kappa_\parallel / \kappa_\perp$

Binding anisotropy: $\beta = \epsilon^b_\parallel / \epsilon^b_\perp$

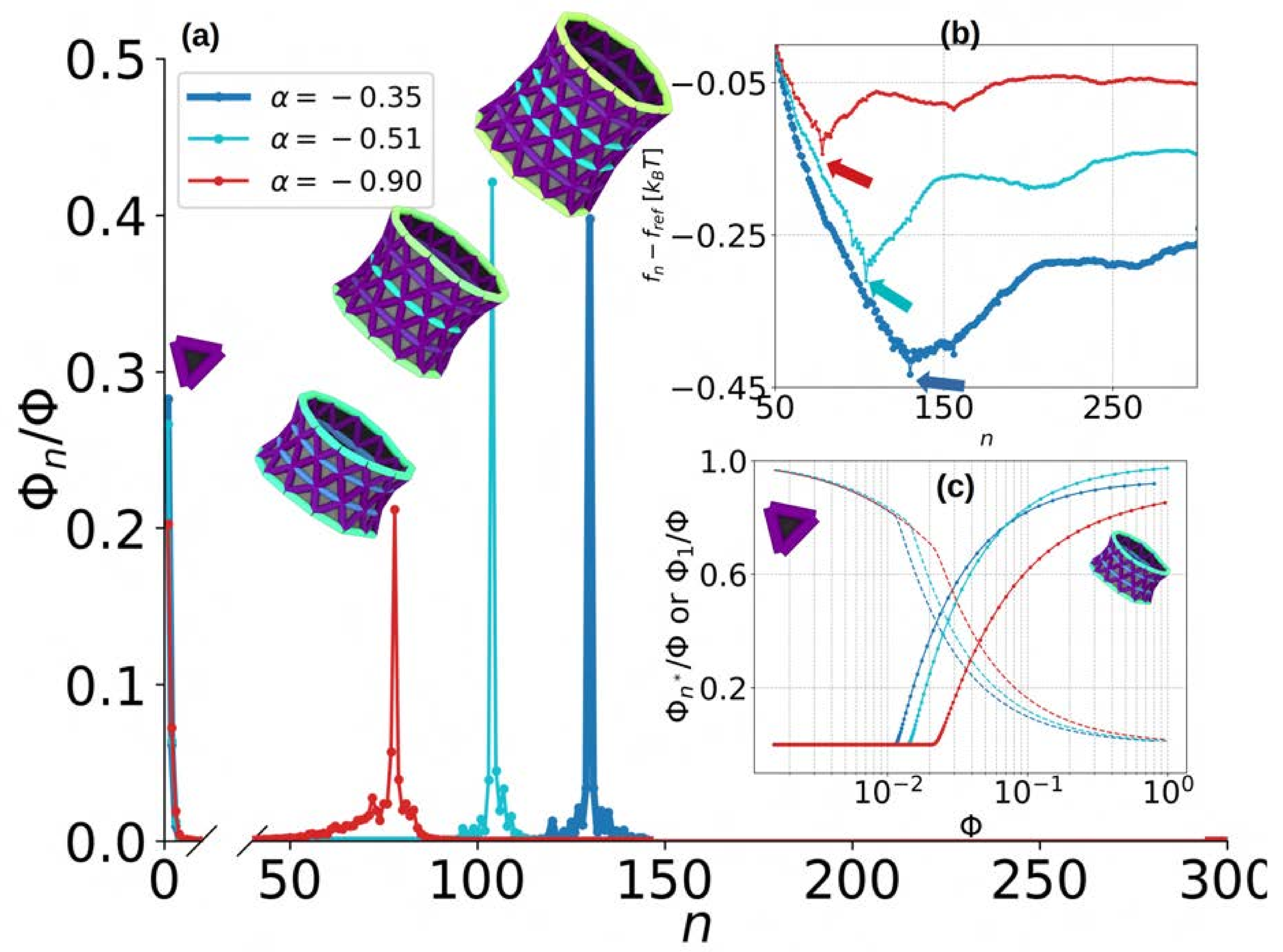
In Grand Canonical ensemble, fixed chemical potential

Results

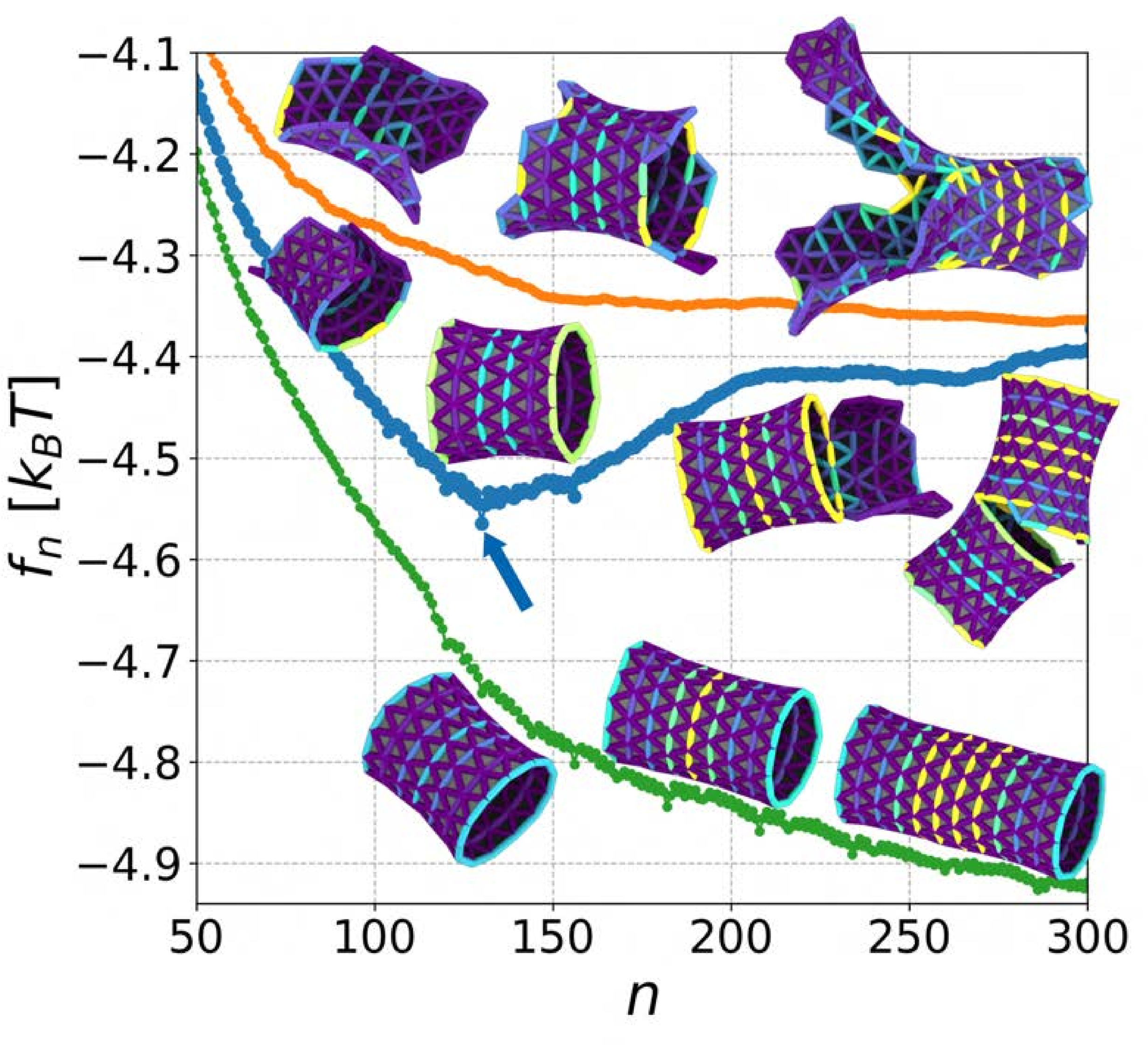
Cluster size growth in time



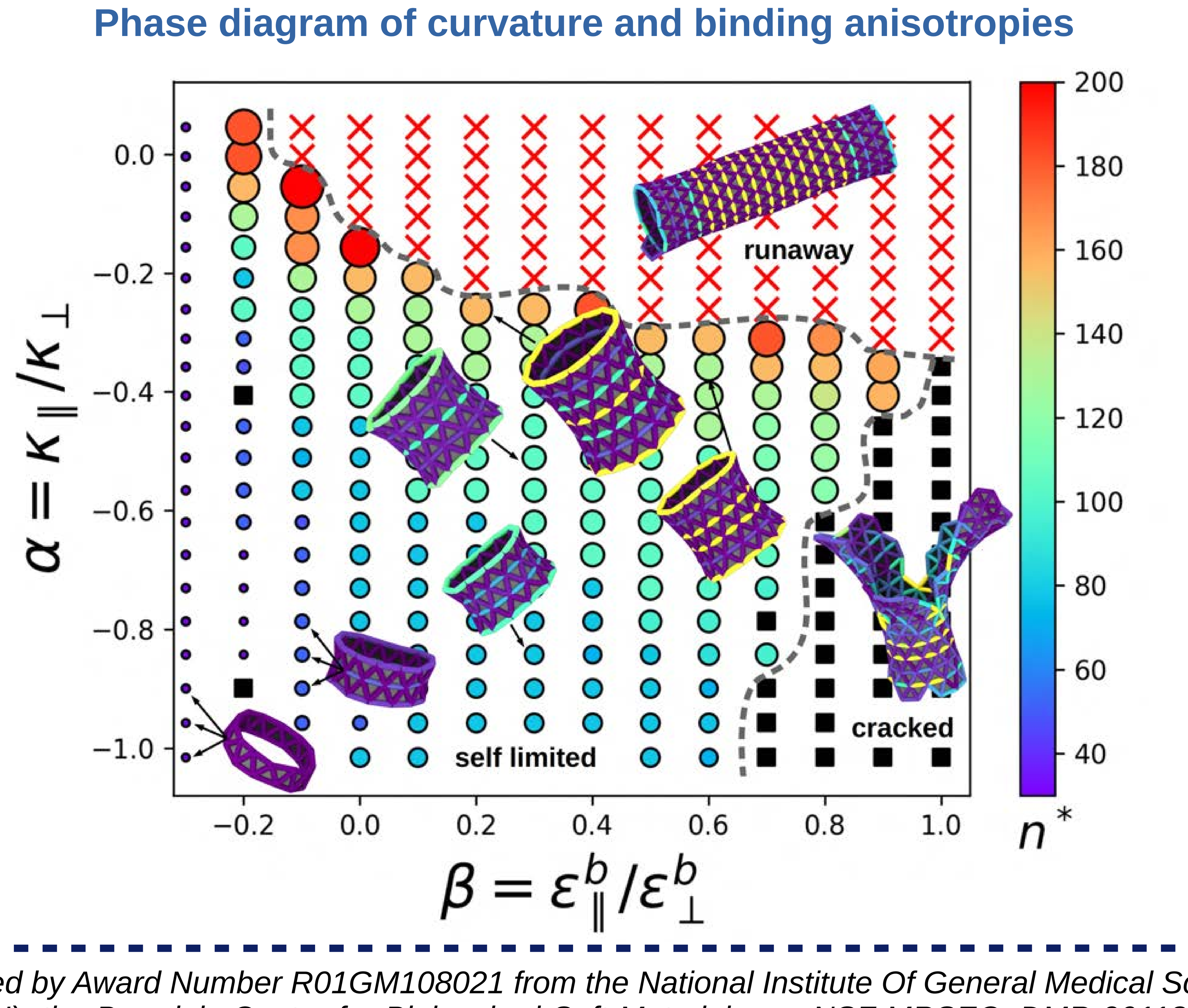
Calculated cluster size equilibrium distributions



Per-particle free energies



Phase diagram of curvature and binding anisotropies



References

G.M. Grason, J. Chem. Phys. 145, 110901 (2016)
G.M. Grason, Nature Physics 13, 1149 (2017)
M.F. Hagan, G.M. Grason, Rev. Mod. Phys. 93, 025008 (2021)
C. Sigl, et al. Nature Materials 20, 1281 (2021)
B. Tyukodi, F. Mohajerani, D.M. Hall, G.M. Grason, M.F. Hagan arXiv:2109.01174 (2021)

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