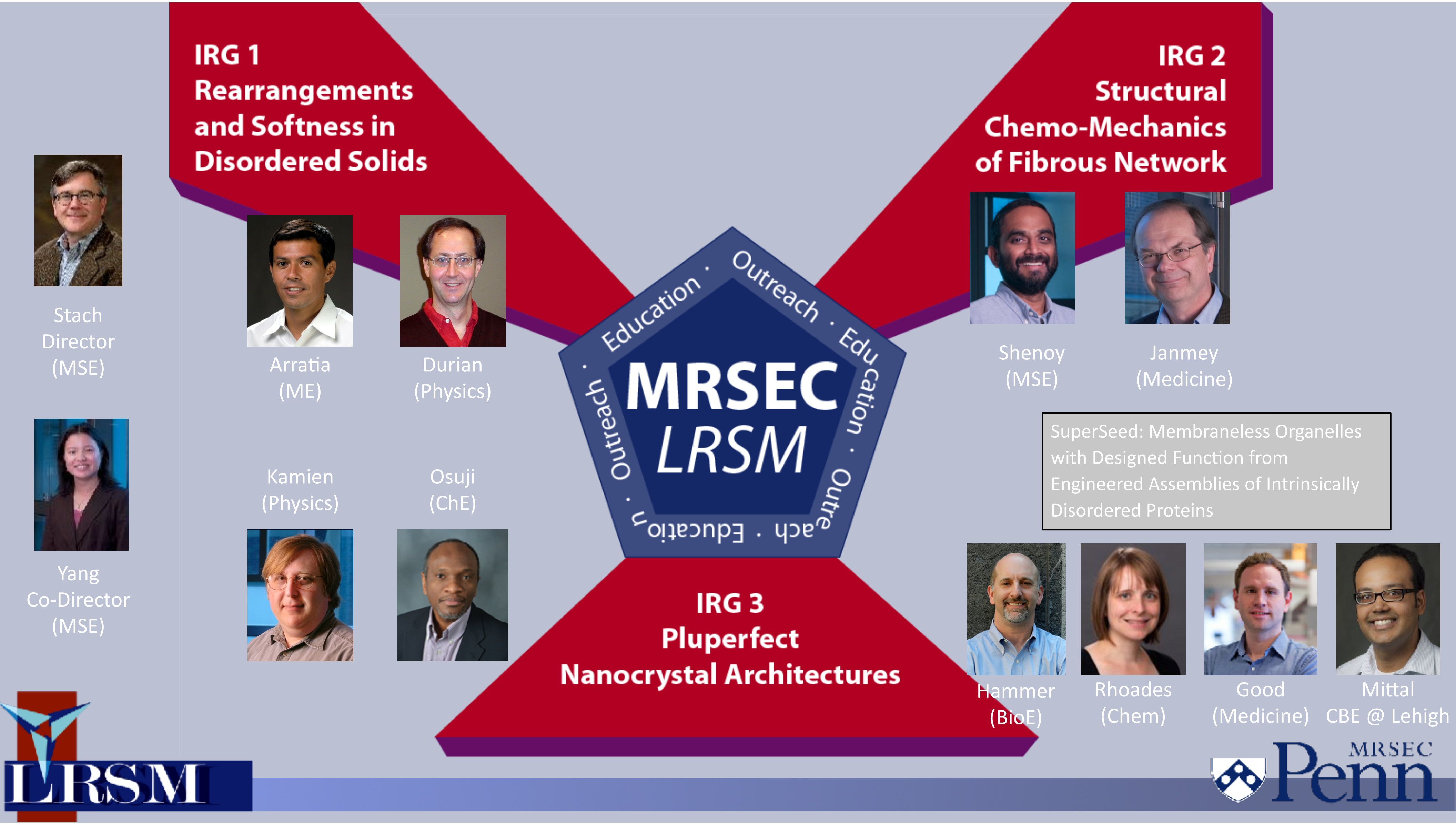

LRSM
Laboratory for Research on
the Structure of Matter


IRG 1
Rearrangements and Softness in Disordered Solids
IRG 2
Structural Chemo-Mechanics of Fibrous Network
IRG 3
Pluperfect Nanocrystal Architectures
MRSEC
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Education · Outreach · Education · Outreach · Education · Outreach
Stach
Director
(MSE)
Yang
Co-Director
(MSE)
Arratia
(ME)
Durian
(Physics)
Kamien
(Physics)
Osuji
(ChE)
Shenoy
(MSE)
Janmey
(Medicine)
SuperSeed: Membraneless Organelles with Designed Function from Engineered Assemblies of Intrinsically Disordered Proteins
Hammer
(BioE)
Rhoades
(Chem)
Good
(Medicine)
Mittal
CBE @ Lehigh
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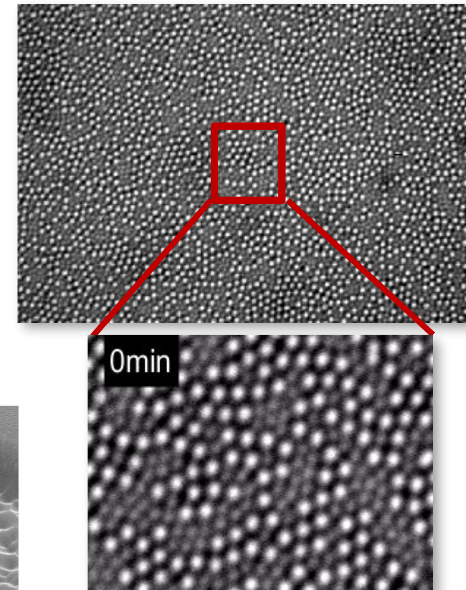
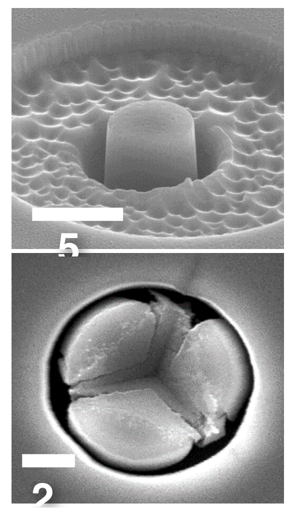
IRG 1: Vision and Goals

Challenge: What material characteristics control flow in disordered solids?

Goal 1: Determine how rearrangements proliferate and interact beyond yield strain

Goal 2: Manipulate cooperative evolution of these rearrangements to widen window between onset of plastic flow and failure by fracture





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IRG 1: “Softness” as a descriptor

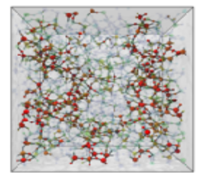
‘Softness’: a structural descriptor identified through machine learning

- Propensity of a given particle grouping to rearrange upon yield
- Analogue to dislocations in crystalline solids

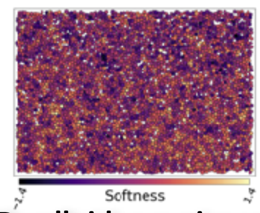
Experiment and theory show that softness captures yield behavior across large length scales

- Disordered solids display common behavior below & near yield
- Spans seven orders of length scale
  - Atomistic to mesoscale

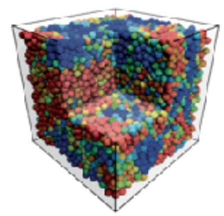
3D SiO_2 simulation
86% CV Accuracy



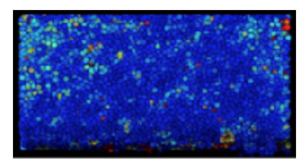
2D colloid experiments
72% CV Accuracy



3D LJ simulation
90% CV Accuracy

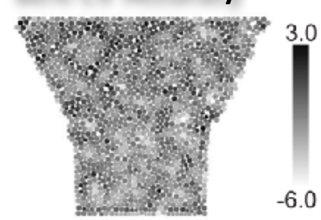


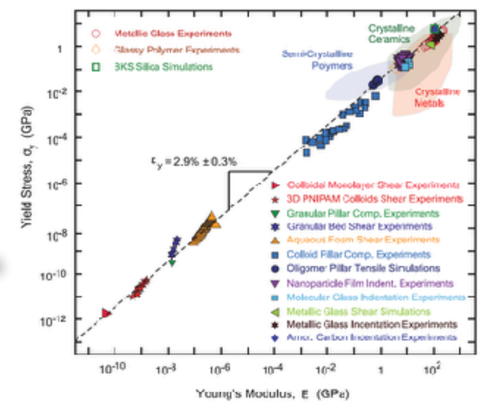
3D colloid experiments
77% CV Accuracy



3D polymer simulation
93% CV Accuracy

2D granular experiments
80% CV Accuracy





Cubuk, … **Arratia**, **Carpick**, **Durian**, **Fakhraai**, **Jerolmack**, **Lee**, **Riggleman**, **Turner**, **Yodh**, **Liu**, Science (2017).

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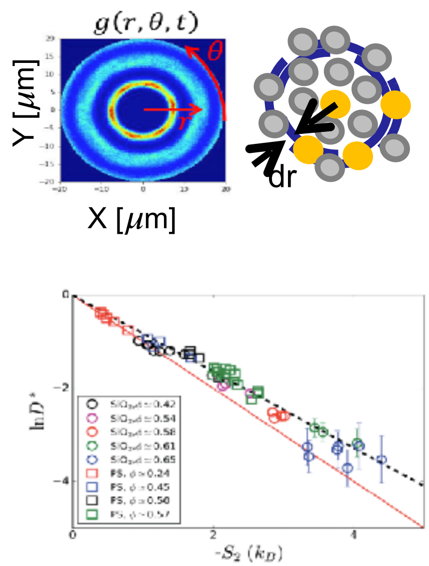
IRG-1: Connecting Structure to Dynamics

"Excess entropy" as measure of disorder compared to ideal gas configuration

It is a structural quantity

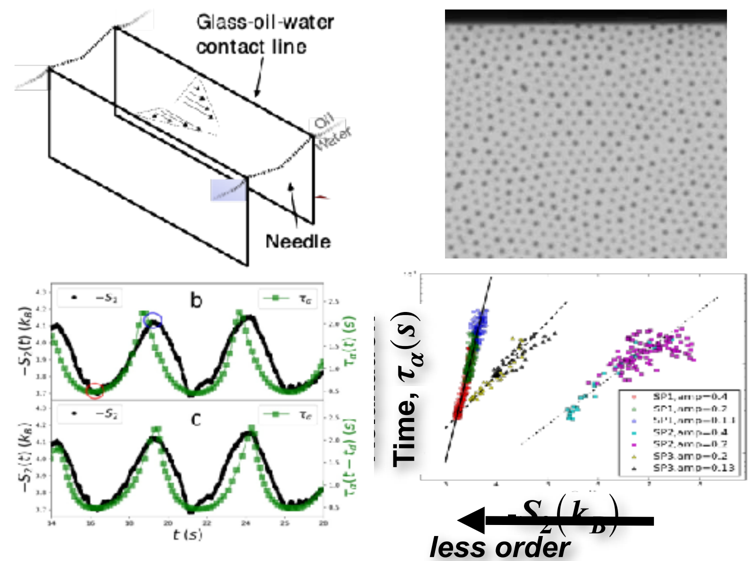
- Can relate long-term particle diffusion to structure
- Microstructure results from relaxation dynamics

Attractive Thermal Colloids



Ma,…**Yodh,** *J. Chem. Phys.*, 2019 (*Editor's Choice*)

Repulsive sheared colloid



Galloway, … **Jerolmack**, **Yodh**, **Arratia**, PNAS, 2020

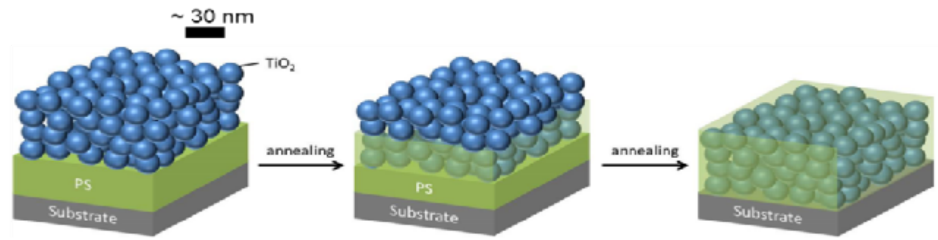
IRG 1: Controlling toughness

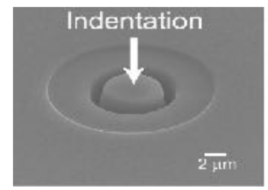
Capillary rise infiltration (CaRI)

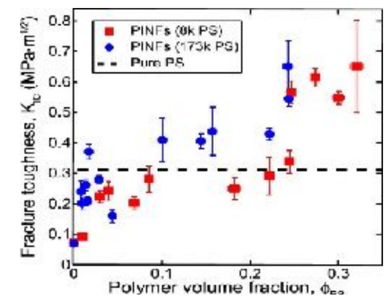
Infiltrate high T_g polymers into nanoparticle packings

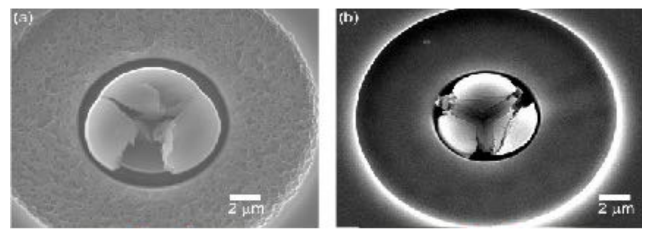
High T_g polymers for glassy, solid bridges between particles

- Increased toughness with increase in polymer volume fraction



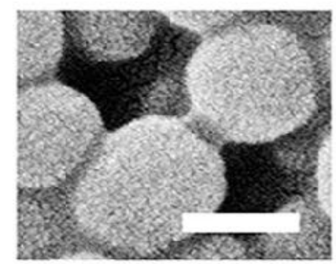






Brittle Failure

Plastic Failure



Hor, … **Riggleman**, **Turner**, **Lee**, ACS Nano, 2017.
Jiang, … **Lee, Turner**, ACS AMI, 2018.
Wang, … **Lee**, **Fakhraai**, ACS Nano, 2018.

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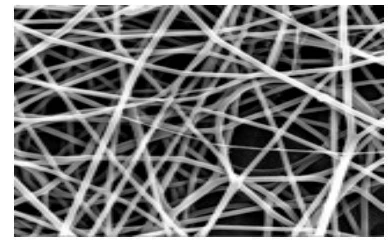
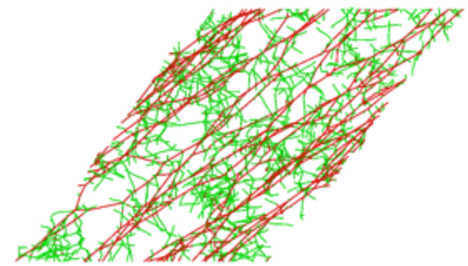
IRG 2: Vision and Goals

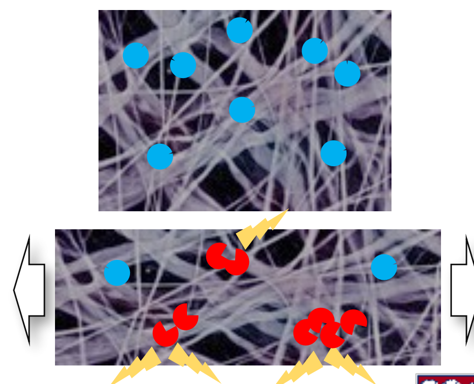
Challenge: Understanding and harnessing structural, chemical, and mechanical complexity



Goal 1. Design and synthesize new fibers and crosslinks



Goal 2. Create and characterize new network geometries for tuned response



Goal 3. Integrate chemo-mechanical function into networks

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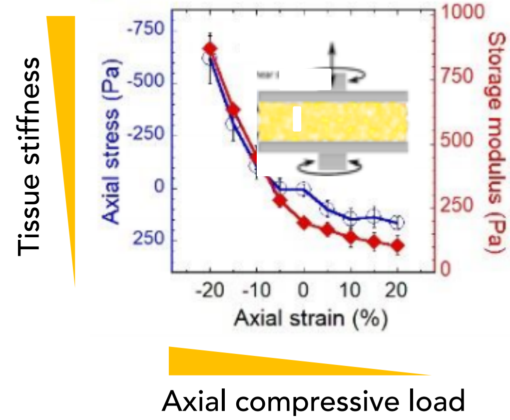
IRG 2: Tissue Mechanics in Fibrous Networks

Experiments on mammalian tissues led to an unexpected experimental finding:

- Shear modulus increases in compression

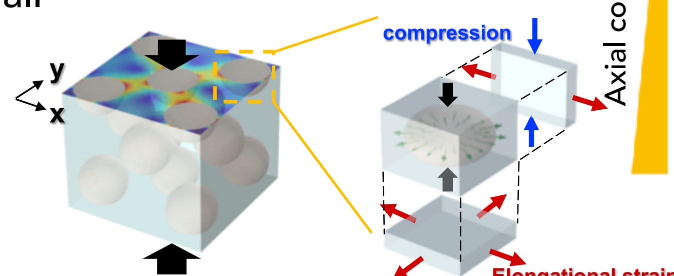


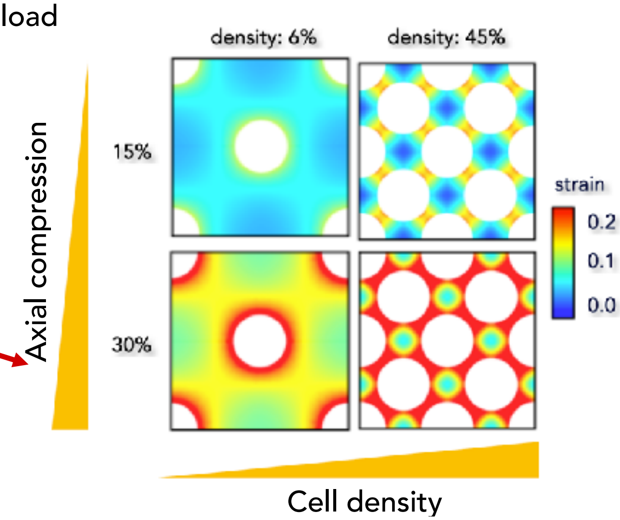
Theory shows that if cells that surround fibrous collagen are incompressible:

- Strain distribution is inhomogeneous
- Leads to tension in the matrix, which translates to compression stiffening overall

Similar observations in plant tissues

- Behavior appears universal





A. van Oosten, … **Shenoy**, **Janmey**, Nature, **573**, 2019

IRG 2: Self-reinforcing fibers

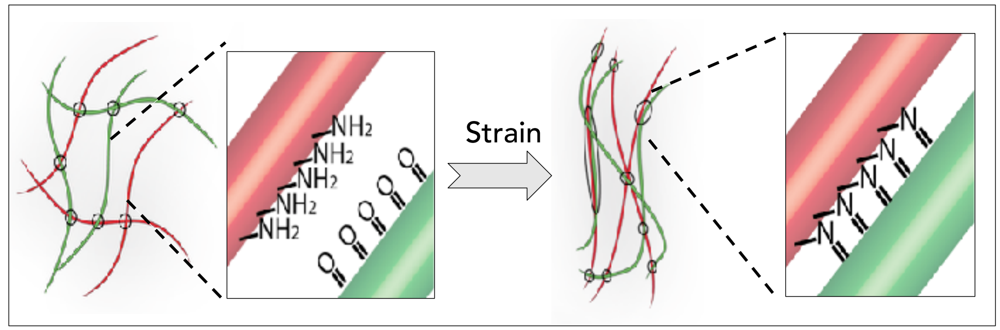
Engineering of variable adhesion strength during electrospinning

- Controls reordering of fiber network
- Leads to control over deformation
  - Moldable
  - Fully reversible

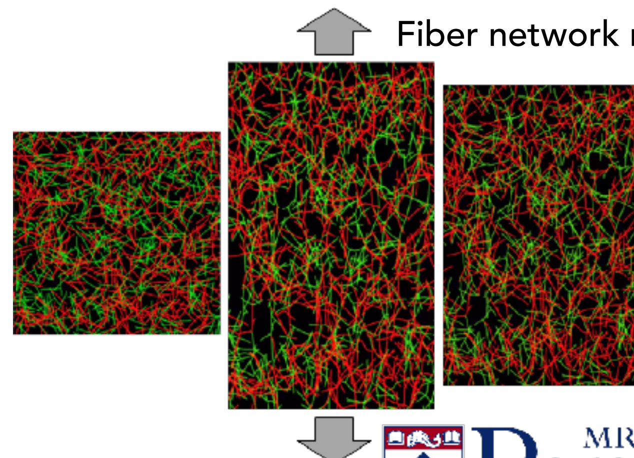
Can be used to guide cell differentiation

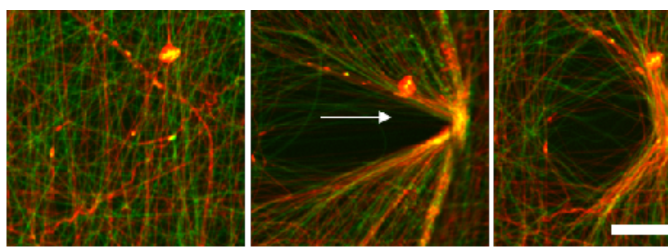
Self-adhesive multi-fiber materials

Mixed fiber populations electrospun with chemical groups that form bonds when brought into contact



Fiber network model

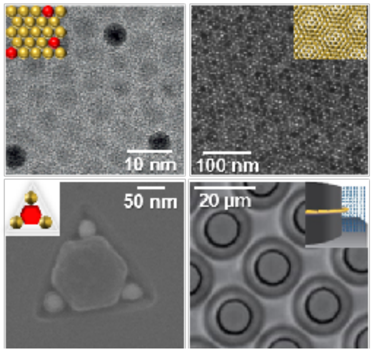




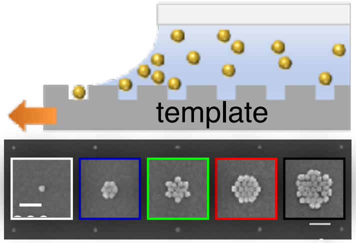
Micro-scale strain measurements track effect of fiber adhesion

IRG 3: Vision and Goals

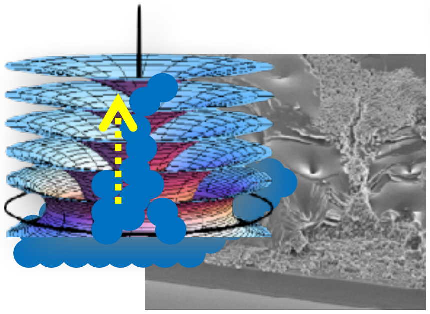
Challenge: How to create nanocrystalline architectures with control over order "beyond perfection"



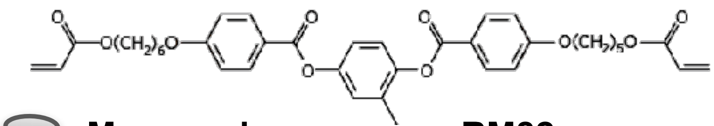
Goal 1. Link nanocrystalline self-assembly and template approaches



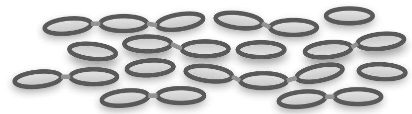
Goal 2. Utilize control over soft matter to guide NC assembly



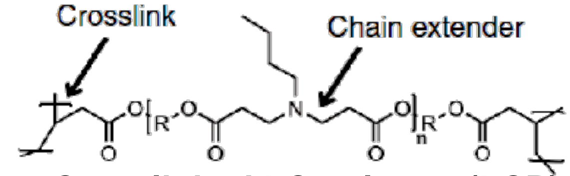
IRG3: Shape transitions in LCO



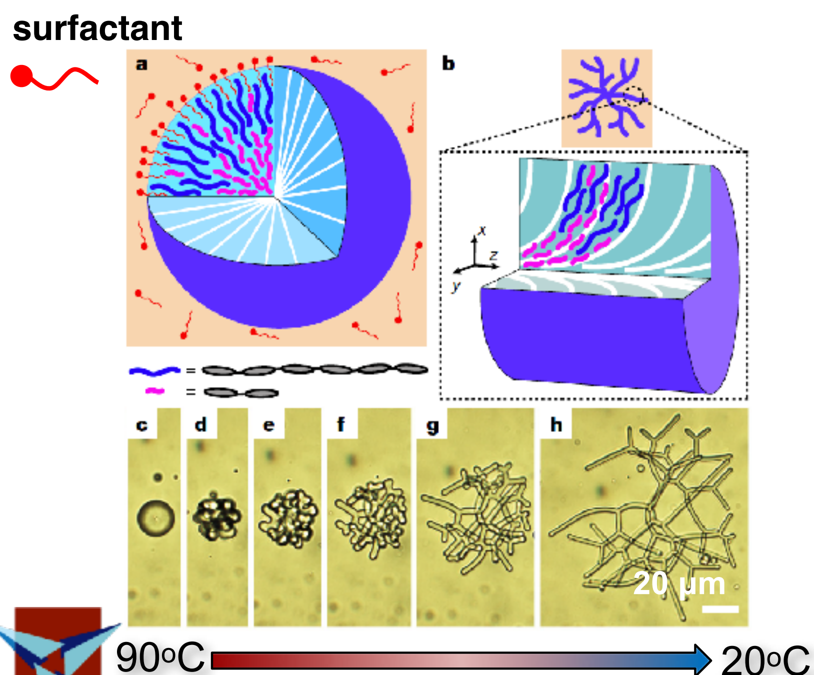
Mesogenic monomer: RM82



Nematic LC oligomer (LCOs)



Crosslinked LC polymer (LCP)

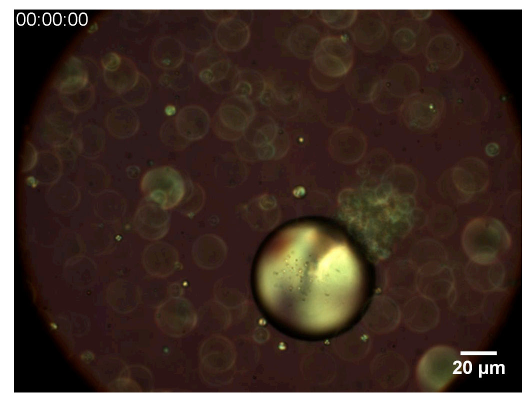


At high temperature (T), simple spherical drops form.

Spherical drops experience spectacular, reversible shape-transitions on reducing T

- Yields filamentous networks

Surfactant concentration, LCO chain length distribution, and T control the shape-transitions.



Depends on delicate balance of LC elasticity and surface tension

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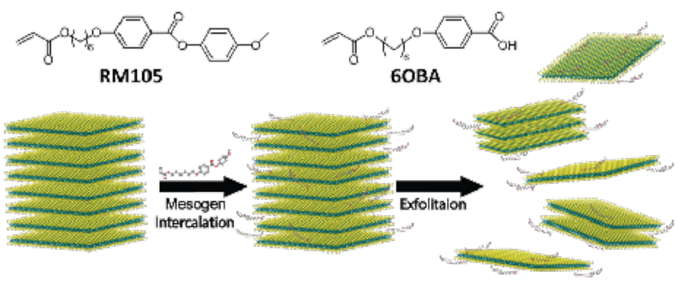
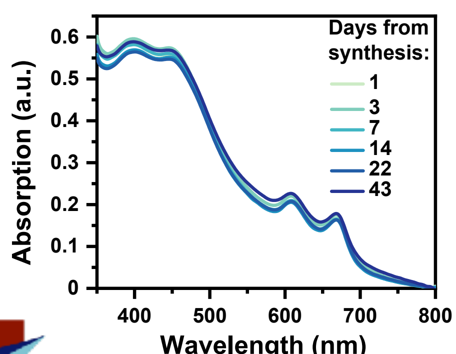
Wei, … **Yang**, **Yodh**, Nature, 2019.

IRG3: Exfoliation & Dispersion of 2D-MoS_2 in LCs

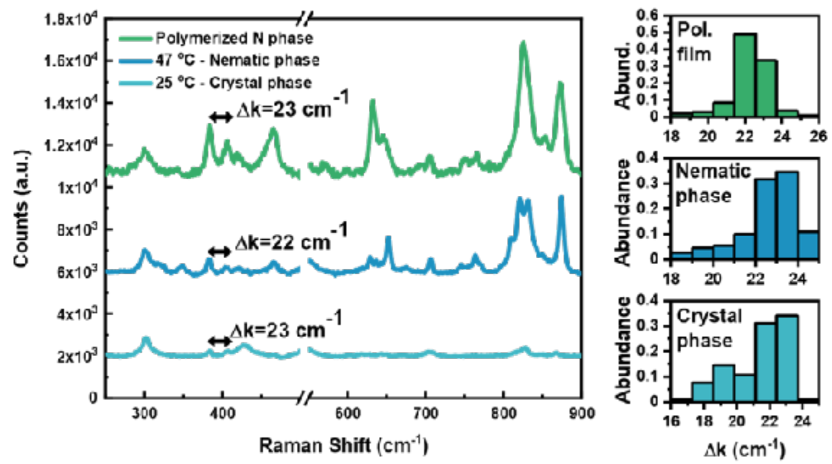
Controlled interactions with mesogens lead to the formation of stable MoS_2 dispersions in solution and thermotropic liquid crystalline phases.



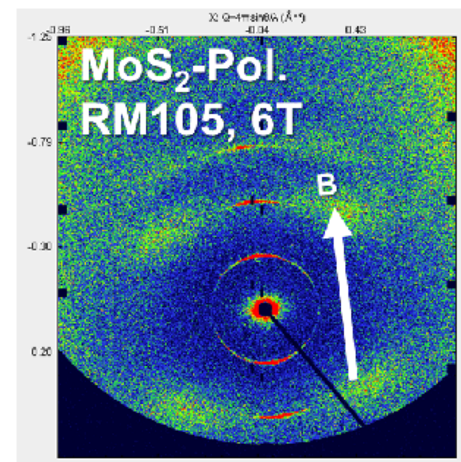
Suspension stability



Stability in LC phases



Magnetic alignment



Stimuli-responsive materials

Insight into anisotropic properties of 2D-MoS_2

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Gabinet, Thompson, **Kagan**, **Osuji**, *et al.* To be submitted (2021).

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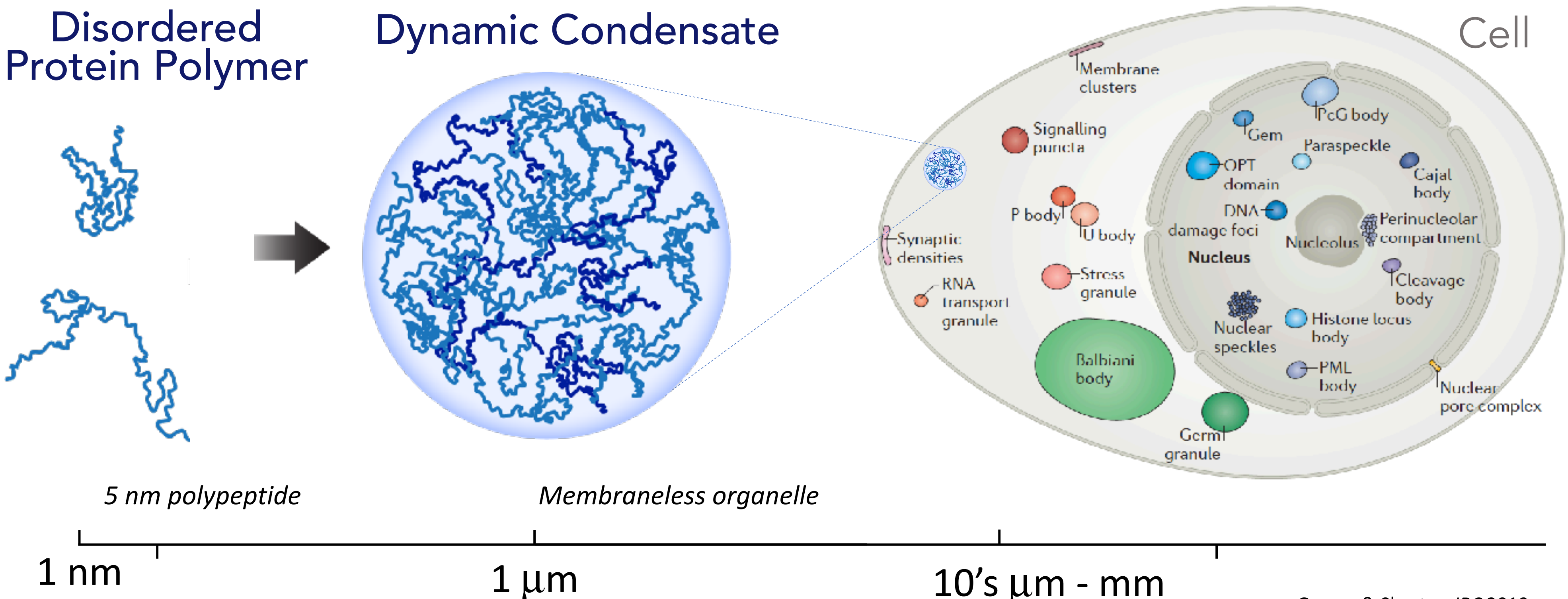
SuperSeed

Self-Assembly of Protein Polymers into Membraneless Organelles



Gomes & Shorter, *JBC* 2019

Banani & Rosen, *Nat Rev Mol Cell Biol* 2017

Liquid-Like Materials that Drive Cellular Organization and Function via Compartmentalization

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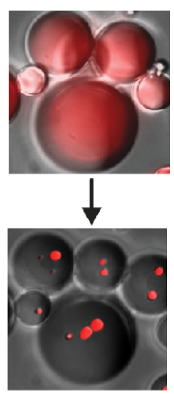
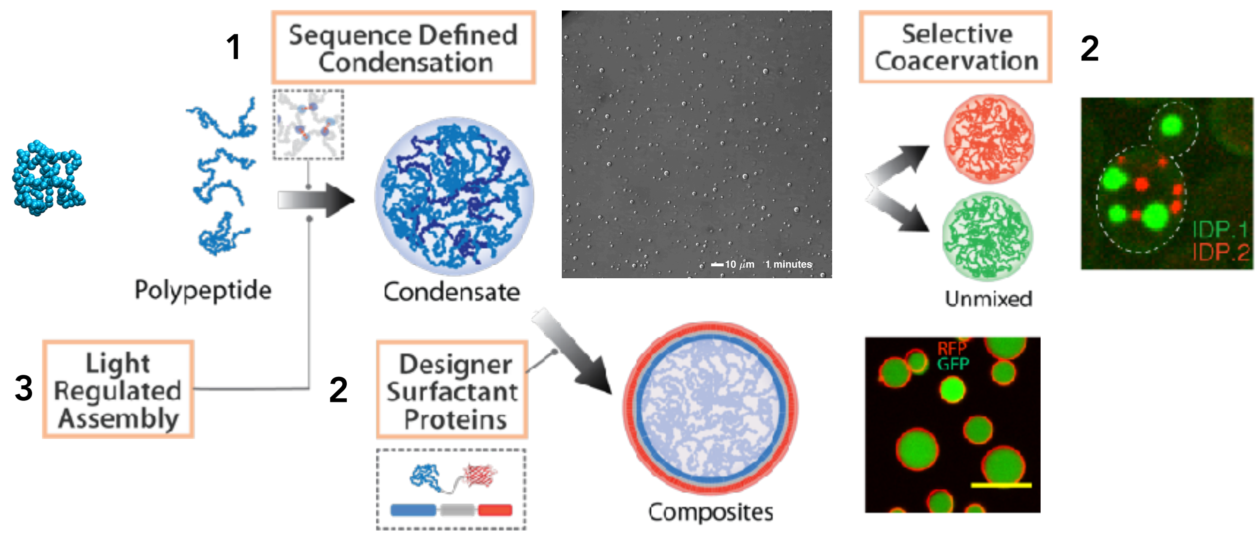
SuperSeed Goals

Novel Materials Design from Coacervating Proteins

Predictive Rules for Phase Separation

Engineered Membraneless Organelles





	Rules of Life	Novel Materials Applications
1	Sequence determinants of IDP phase separation	Designer coacervating materials; tunable properties
2	Fidelity of protein condensation in a living system	Orthogonal protein compartments in cells
3	Membranelles organelle function	Control of cell behavior through synthetic membranelles organelles

Team: Theory and Experimental Expertise

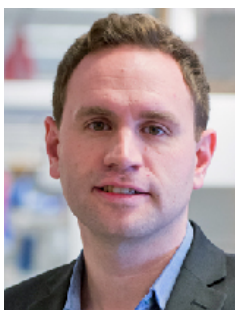
Liz Rhoades

Chemistry
IDP Biophysics

Dan Hammer

Bioengineering
Protein Engineering

Matt Good

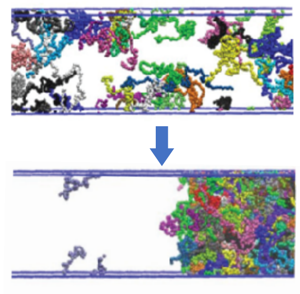


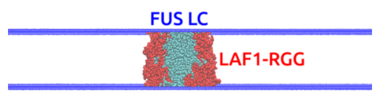
Cell & Dev. Biol.
Synthetic and Cell Biology

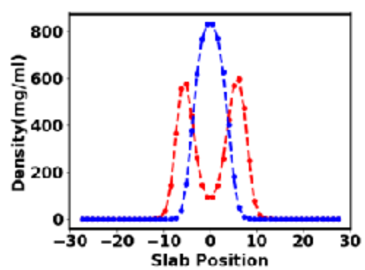
Jeetain Mittal

Chem. Biol Eng.
Computational Modeling

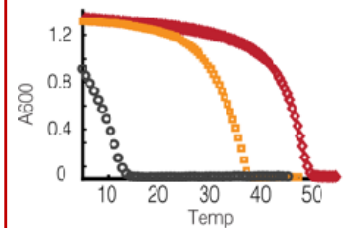
Coarse grained Simulations

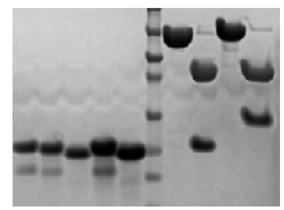


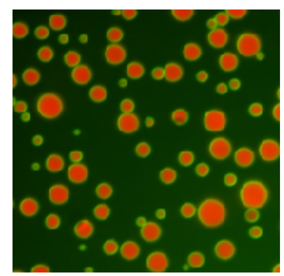




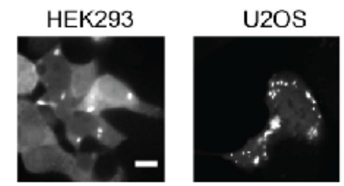
Protein Synthesis, Characterization

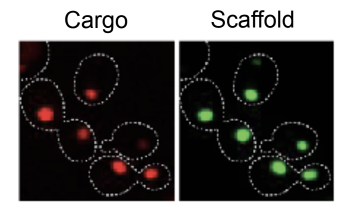


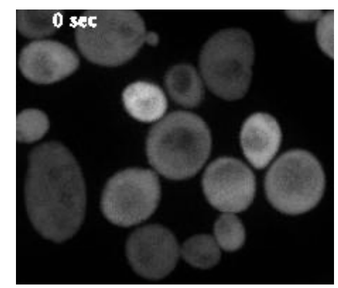




Materials Engineer. Of Living Cells







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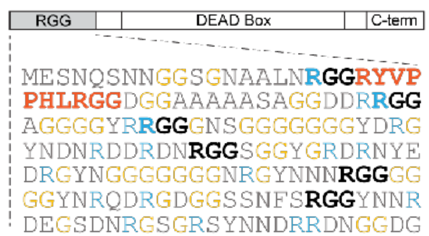
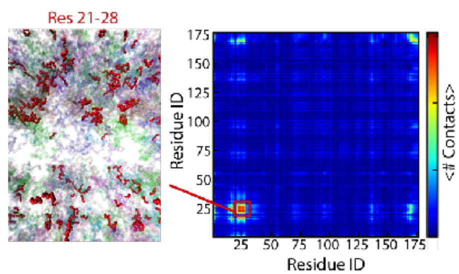
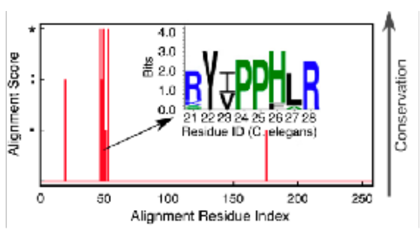
Highlight #1 : Sequence Determinants of IDP Coacervation

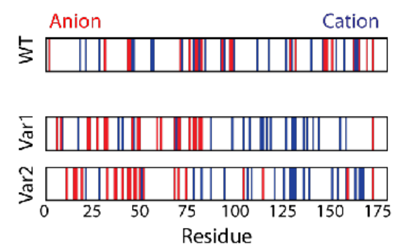
Identify Sequence Rules



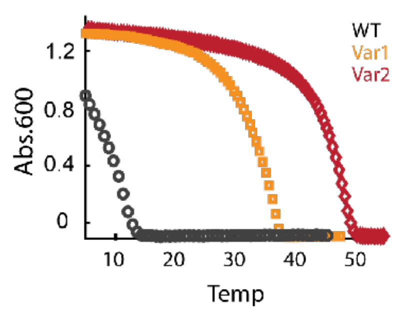




Design of New Sequence

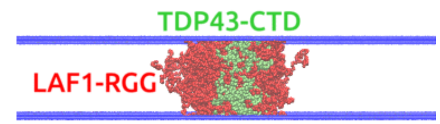


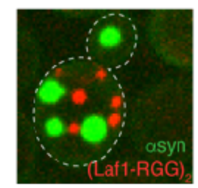
Tunable LLPS

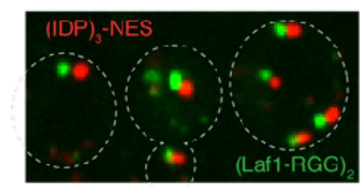


Schuster et al, *PNAS* 2020
Schuster et al, *Nature Comm* 2018
Dignon et al, *Plos Comp. Biol.* 2018

Selective Coacervation







Garabedian et al,
manuscript in prep 2021

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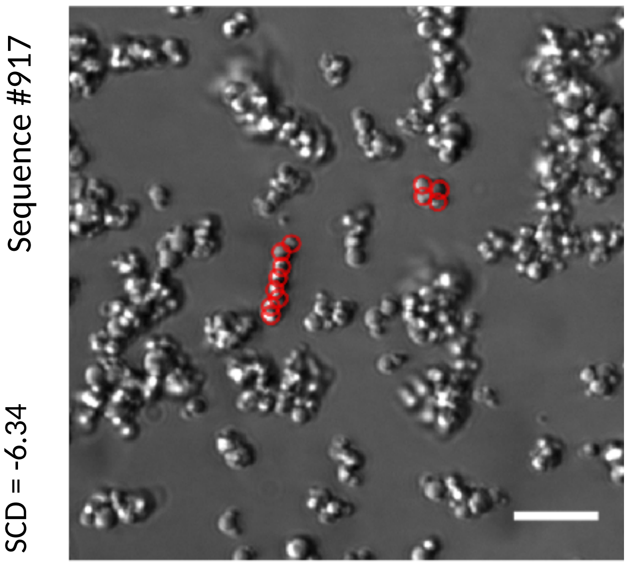
Highlight #2 : Engineered Interface Properties

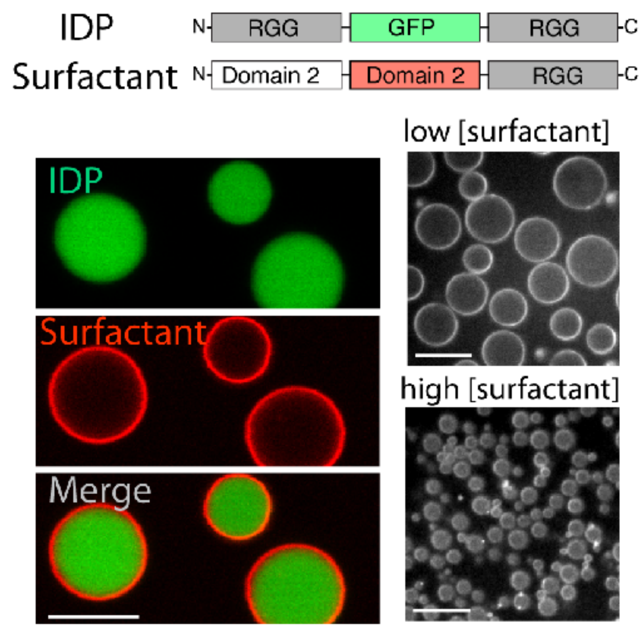
Sequence Design

MDEAEDGSGGNNGNGSGYGGRDYDGDGGGNGGSYA
DNGDGRDGDDGGAGGNGSGSDGGDNPDDGLRGRGR
GNGGFGNGNRGGSREGGGRGYSDNGNSHGDNNRSP
AGQAYGSDARRSGRRGGRNNGSNRNRGRYGRGQNY
YLRYGRAYNGRGRGNGANRARYGNGVGGLE



Droplet Fusion Blocked

Surface Active Agents



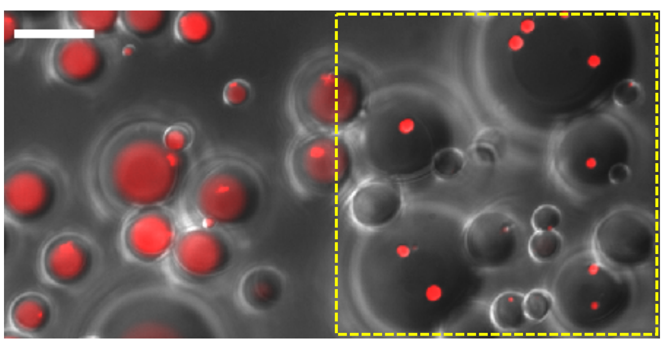
Composites of tunable size

Highlight #3: Light-Responsive IDP Materials

Stable Protein Coacervation Using a Light Induced Transition

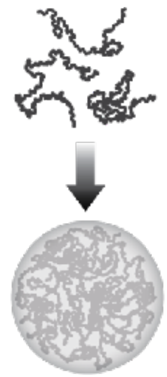
Dark

Illuminated (405 nm)

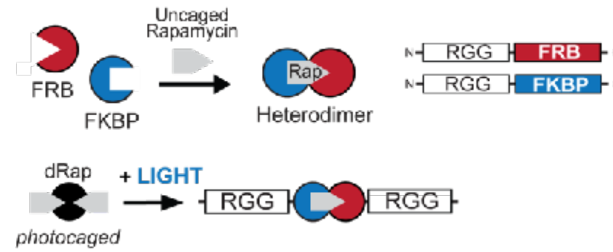


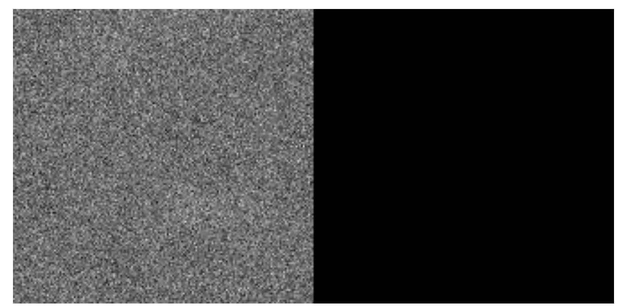
Single Pulse; Condensates stable over time

Reed et al, *ACS Syn Biol* 2020



Valency Increase for Stable Coacervation of IDPs





Garabedian et al, *under revision, Nature Chem Biol* 2022

IRG 1
Rearrangements and Softness in Disordered Solids

Fluctuations as a Microscope for Characterizing Rheological Material Behavior

Dynamic Behavior of Random Media in the Absence of Scale Separation

Molecular Organometallic Sorbents with Tunable Magnetic Materials Properties

Packaging and Release of mRNA and of other Macromolecules from Supramolecular Virus-Like Assemblies

IRG 2
Structural Chemo-Mechanics of Fibrous Network

Sequence-Defined Disordered Protein Polymers for Engineered Assembly of Biomolecular Condensates and Granular Materials

Materials from Disordered Bicontinuous Aperiodic Networks (D-BANs)

Nonlinear and Nonequilibrium Topological Materials

Imaging and Manipulation of Spin-Textures and Magnetic Phase Transitions in Atomically-Thin Van der Waals Materials

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IRG 3
Pluperfect Nanocrystal Architectures

Efficiently exploring high-dimensional energy landscapes in experiment and computation

Ultrafast Measurements of Interface Thermal Conductance using Dye-Based Optical Thermometry

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