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**Grand Challenges in Soft Matter Science: Prospects for Microgravity Research**

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**Active patterning and structure formation – self-limiting assembly, actuation, and integration**

Primary Author:

William Meyer, NASA Glenn Research Center  
216-433-5011  
William.V.Meyer@nasa.gov

Co Authors:

Christian Santangelo, Syracuse University  
Karen Daniels, NC State University  
Boris Khusid, New Jersey Institute of Technology University  
Arjun Yodh, University of Pennsylvania  
Bhuvnesh Bharti, Louisiana State University  
Stuart Williams, University of Louisville  
Taylor H. Ware, Texas A&M University  
Robert W. Cohn, University of Louisville  
Douglas Durian, University of Pennsylvania  
Robert Leheny, Johns Hopkins University  
Travis W. Walker, South Dakota School of Mines & Technology  
Endao Han, Princeton University  
Xiang Cheng, University of Minnesota  
Alice Quillen, University of Rochester  
Wylie Ahmed, California State University Fullerton  
Mohamed Amine Gharbi, University of Massachusetts Boston  
Joanna Robaszewski, Harvard University  
Peter Olmsted, Georgetown University  
Margaret Gardel, University of Chicago  
Stefano Martiniani, University of Minnesota  
Michelle Driscoll, Northwestern University  
Jeremy Schmit, Kansas State University  
Samantha McBride, Princeton University  
Abdul Malmi Kakkada, Augusta University  
Lou Kondic, New Jersey Institute of Technology  
Lorena Tribe, Penn State Berks  
Zvonimir Dogic, University of California at Santa Barbara  
Nathan Keim, Pennsylvania State University

Jarrold Edward Schiffbauer, Colorado Mesa University  
Matthew Lynch, Procter & Gamble Company  
Alicia Boymelgreen, Florida International University  
Vikram Jadhao, Indiana University  
Catherine G. Reyes, Lawrence Livermore National Laboratory  
Charles Schroeder, University of Illinois at Urbana-Champaign  
M. Lisa Manning, Syracuse University  
William Irvine, University of Chicago  
Peter Schall, University of Amsterdam  
Chang-Soo Lee, Chungnam National University  
Joe MacLennan, University of Colorado Boulder  
Noel Clark, University of Colorado  
James Ferri, Virginia Commonwealth University

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# Grand Challenges in Soft Matter Science: Prospects for Microgravity Research

*Paul Chaikin, Compiler*  
*New York University, New York City, New York*

*Noel Clark*  
*University of Colorado Boulder, Boulder, Colorado*

*Sidney Nagel*  
*University of Chicago, Chicago, Illinois*

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Mail Stop 148  
NASA Langley Research Center  
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NASA/CP-20205010493



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*Paul Chaikin, Compiler*  
*New York University, New York City, New York*

*Noel Clark*  
*University of Colorado Boulder, Boulder, Colorado*

*Sidney Nagel*  
*University of Chicago, Chicago, Illinois*

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Grand Challenges in Soft Matter and Opportunities for Microgravity Research at  
2020 American Physical Society (APS) March Meeting  
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National Aeronautics and  
Space Administration

Glenn Research Center  
Cleveland, Ohio 44135

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## Executive Summary

The worldwide community of soft matter has grown rapidly in size, impact, and scope since the last NASA report in 2003 as evidenced by the organization of the new Division of Soft Matter (DSOFT) at the American Physical Society, and the arrival of a dedicated international journal “Soft Matter” published by the Royal Society of Chemistry. At the suggestion of NASA’s Physical Science Research Program in the Space Life and Physical Science Research and Application Division<sup>1</sup> for an update of the 2003 report on the NASA Soft and Complex Condensed Matter Workshop, Paul Chaikin, Noel Clark, and Sidney Nagel, organized a focus session and workshop for the 2020 American Physical Society (APS) March meeting under the auspices of the DSOFT. Due to the COVID-19 pandemic, the March meeting was canceled and the workshop “Grand Challenges in Soft Matter and Opportunities for Microgravity Research” was reincarnated as a remote Zoom (Zoom Video Communications, Inc.) meeting convened Thursday, March 26, 2020, from 11:30 a.m. to 1:30 p.m. EST. After a brief introduction, the ~100 participants (mostly from the United States with several joining from the European Union) separated into eight breakout sessions on

1. Self-organization only possible far from equilibrium—machines making machines
2. Instrumentation—from neuromorphic computing to large-scale self-assembly
3. Suspensions, foams, emulsions, colloids, and granular materials—self-healing, tuning gravity, and life support for exploration
4. Packings, simulation, and big data—artificial intelligence emulation of soft matter
5. Mechanical metamaterials and topological soft matter: allostery and auxetics—distributed energetics and mutation upon deployment
6. Soft matter, bioscience, and biotechnology—evolution and the marginal stability of life
7. Active patterning and structure formation—self-limiting assembly, actuation, and integration
8. Fluids: liquid crystals—self-assembly of the superlarge and superweak active clothing

The participants then reassembled for a presentation of conclusions and general discussion. Three overarching themes emerged from this workshop and are presented with additional details:

- Machines made out of machines
- Scalable self-sustaining ecosystems
- Active materials and metamaterials

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<sup>1</sup>Space Life and Physical Science Research and Application Division has moved to the Science Mission Directorate and is now the Biological and Physical Sciences Division.





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# Grand Challenges in Soft Matter Science: Prospects for Microgravity Research

Paul Chaikin  
New York University  
New York City, New York 10003

Noel Clark  
University of Colorado Boulder  
Boulder, Colorado 80309

Sidney Nagel  
University of Chicago  
Chicago, Illinois 60637

## Abstract

At the suggestion of NASA's Physical Science Research Program in the Space Life and Physical Science Research and Application Division, Paul Chaikin, Noel Clark, and Sidney Nagel organized a focus session and workshop for the 2020 American Physical Society (APS) March meeting under the auspices of the Division of Soft Matter.

Three overarching themes emerged from the workshop and are presented with additional details:

- Machines made out of machines
- Scalable self-sustaining ecosystems
- Active materials and metamaterials

This report lays out only some of the potential directions for soft matter dynamics over the next two decades. It also lays out the role that gravity plays in the organization of the basic building blocks of matter. Not only will research on soft matter have tremendous application towards understanding its behavior in our terrestrial environment, but also potentially in other NASA programs such as planetary science, exploration, robotics, etc.

## 1.0 Introduction

There was great enthusiasm for the breakthroughs that have occurred in the past two decades: in materials, in experimental, simulational, and theoretical techniques, and in technologies and phenomena discovered. As has been emphasized in other reviews of the soft matter research challenges (Nagel, 2017), these advances have significantly changed the vision and direction of this field and portend an exciting future.

There was a great deal of overlap in the goals, directions, and challenges that arose within the separate breakout sessions:

1. Advances in machine learning, artificial intelligence, big data, and simulations have changed not only the way data are currently analyzed but also affect how experiments are being conceived and executed.

2. New material building blocks have been developed. These include designer particles and materials with activity that have the ability to sense their surroundings, shift shape, communicate, self-assemble, self-replicate, and respond specifically at remote distances (e.g., as in allosteric proteins).
3. New symmetries, symmetry breaking, and topological protection have been discovered.

Taken together these advances allow us to tackle more complex fundamental problems that involve far-from-equilibrium phenomena, complexity, and cooperativity and to imagine lifelike materials and machines of technological importance. In Section 1.2, we highlight three overarching themes that have emerged from this workshop.

## 1.1 Complex Is Different

The field of soft matter began with the study of materials built not with simple atoms or molecules as building blocks but with larger entities, such as polymers, colloids, nematogens, or surfactants, all of which have many internal degrees of freedom. These are the molecules and components of life. Consequently, the cross-fertilization between soft matter and biology has been extensive. As the field has evolved, the ingenuity of chemists has imbued soft materials with more lifelike properties. The building blocks now have properties such as activity; recognition capability; chemical, temperature, or light sensitivity; and the ability to change shape. One theme that arose in the workshop was why not go one step further and let them “think” and communicate? A basic question then is how to understand, control, and use such a complex dynamical system so that it does not destroy itself chaotically.

This is also the aim of much of 21st-century science. While the 19th and 20th centuries effectively solved the problem of equilibrium thermodynamics and statistical mechanics, the work of this century will be to develop an equivalent understanding of nonequilibrium statistical mechanics, phenomena, and dynamics. As a way to tackle these essential fundamental and technological problems, we suggest the following grand challenges.

## 1.2 Grand Challenges—Toward Artificial Life

Creating artificial life has many grand challenges including

1. Machines made out of machines: Every living thing is a machine made of machines (Needleman and Zvonimir, 2017). One goal is to create materials that behave in the same way. Imagine an interacting network of active elements each with its own power source and each connected to its neighbors chemically or physically. What we know from network theory and from preliminary experiments is that interesting phenomena occur as soon as the network nodes are nonlinear. We expect such machines to be able to self-repair, self-replicate, microcompartmentalize, specialize, operate as distributed engines, compute, and learn. Aside from the clear utility of intelligent machines, they also demand an understanding of complex dynamics and cooperativity that we sorely lack. These machines would have the potential to impact technologies from smart fabrics to soft robotics.
2. Scalable self-sustaining ecosystems: Systems that recycle their building material using only an external power source are found in cells and on the planetary scale. With the advent of three-dimensional (3D) printing of structures, one challenge is to create such devices and clothing that can be recycled, for example, during a space voyage. As an example, biology uses metastable polymers that can draw energy from themselves. Designing self-sustaining ecosystems with

similar properties has become a major challenge. Processing materials is intrinsically nonequilibrium and requires understanding rheology, friction, and dynamical self-assembly in systems that are glassy, granular, disordered, or suspended particulates. This challenge aims at understanding fundamental dynamical organizational principles and advancing technologies such as hierarchical additive manufacturing, recycling, conservation, and pharmaceuticals and biomedical applications.

3. Active materials and metamaterials: Attractive active particles can condense into a liquid phase with unusual properties that are only beginning to be understood or explored. They can be engineered to have negative viscosity—they output power into useful work rather than intake power and dissipate it. Spinning particles and other parity-violating systems that break time-reversal symmetry give rise to dissipationless “odd” viscosity coefficients. New types of order and phase transitions are dynamically accessible as in active liquid-crystal phases. The elasticity of active solids is even less explored. It entails the presence of “odd” elastic moduli absent in continuum theories constructed from energy minimization. Metamaterial realizations of active solids have already revealed exciting macroscopic optical, magnetic, and mechanical properties induced by the interplay between designed microscopic structure and activity.

In addition, other topics that were discussed include the following:

- Intelligent processing and machine learning: The pursuit of such challenges requires new ways of doing science. The explosion in the amount of data that is now generated from experiments and evermore-sophisticated simulations requires intelligent processing. Machine learning is used not only in analyzing experiments and simulations but in designing and running experiments, recognizing, categorizing, and discovering new phenomena, and even formulating theories. Experiments may in turn inform an understanding of how machine learning works.
- The role of microgravity: Dissecting complex phenomena requires isolating individual phenomena, activity, and function, and building up the knowledge of how many parts work together. Gravitational sedimentation limits the experimentalist to two dimensions or systems with gradients. Gravity subjects the systems to external uncontrolled forces and enhances the often-unwanted effects of friction and transport due to thermal convection. Active processing in microgravity will differ from that on Earth and we must study both environments to control each separately.
- Instrumentation that would facilitate such dynamic nonequilibrium experiments would involve modification of present microgravity experiments to allow magnetic, electric, and optical fields flow, shear, and other activation, some of which is already implemented in the European Space Agency (ESA) live-cell imaging in space (FLUMIAS) and Soft Matter Dynamics experiments. Modifications of the magnetic sample mixing presently on the International Space Station (ISS) would allow rotation of magnetic colloids in the Liquid Crystal Facility and the production of metafluids with odd viscosity; adapting the sample capillaries to microfluidic devices would allow controllable hydrodynamics. Microfluidic, droplet microfluidic, and other high-throughput devices would also allow present or modified microscopes to process thousands or millions of samples rather than a few. Microscope modifications that allow laser tweezing will allow different forms of activation and also be useful for biological samples. The use of high-speed cameras will enable new sets of dynamical experiments. Many of these experiments will require massive high-speed data acquisition, transmission, and storage. These new or modified instruments provide a gateway toward our grand challenges.

## 2.0 Breakout Sessions

### 2.1 Self-Organization Only Possible Far From Equilibrium—Machines Making Machines

#### 2.1.1 Self-Assembly

Equilibrium self-assembly, the growth of crystals, alloys, colloidal, and polymer structures, has been known for thousands of years and understood fundamentally over the past two centuries from the development of statistical mechanics. Potential energies and entropy together provide us with thermodynamic potentials that through diffusive motion, guide particles to configurations in which these thermodynamic potentials are minimized. The materials and structures formed can be programmed from the specific interactions of the particles and lead to static configurations.

Nonequilibrium self-assembly involves forces rather than potentials and there are as yet no general principles such as free-energy minimization that can be applied, just Newton's (or quantum mechanical) equations of motion. But the processes yield new ways of making not just static structures but active devices and dynamical systems that can move and morph, transform energy, and even perform the functions of life. The forces can derive not only from potentials but from chemical activity and flow like hydrodynamic interactions and motors. Understanding nonequilibrium phenomena is a fundamental undertaking of 21st-century science and technology. Modern material processing is predominantly far from equilibrium and new types of driven self-assembly allow the creation of materials and devices not previously possible.

#### 2.1.2 Active Matter

Motility-induced phase separation (MIPS) (Cates and Tailleur, 2015) is a beautiful example of dynamic self-assembly. Active matter, fish, birds, bacteria, and artificial colloidal swimmers, can form dense clusters, flocks, and even crystals, not because they attract one another but because as they swim, they slow down when they are crowded together creating a net inward flux. In the past two decades, chemists, physicists, and material scientists have produced a wonderful collection of active particles including chemically fueled phoretic swimmers and spinners, magnetic swimmers and rollers, light-activated and fueled particles, surface-tension driven Marangoni swimmers, electrical field-driven (induced-charge) electrokinetic, and Quincke spinners (that make use of spontaneous electrorotation of a dielectric sphere submerged in a conductive fluid exposed to a static electric field) (Gompper et al., 2020; Bricard et al., 2013).

Spinning particles can form a fluid with “odd” viscosity (Banerjee et al., 2017) where you shear the fluid and it densifies or ping it and you get a new type of sound. Rotating particles can create flows that trap one another in a dynamic motile cluster, again with no potential attraction (Driscoll et al., 2017). Externally driven systems also present opportunities. Sheared colloids self-assemble into a hyperuniform phase not permissible in equilibrium but with useful photonic properties (Wilken et al., 2020). Elongating particles spontaneously choose a handedness and circulate clockwise or counterclockwise (Wu et al., 2017). When you use internally or externally driven systems, you break all sorts of equilibrium theorems and untie many restrictions.

#### 2.1.3 Prospects

We have seen some examples of what can be done with active particles that interact with simple forces: they self-assemble into more complex and interesting entities. They are micromachines making machines on a larger scale. This resembles what we find in living systems as, for example, molecular motors in organelles or cells in organisms. Every living thing is a self-assembled machine made out of

machines (Needleman and Zvonimir, 2017). What we need are more and different kinds of interactions between our particles, communication, sensing, and responding. The communication can be chemical, optical, and acoustic. Some of these ideas have been demonstrated on a centimeter scale in miniature robotics where individual robots, kilobots, with simple instructions and some communication can assemble a myriad of dynamic structures (Rubenstein, Cornejo, and Nagpal, 2014).

We aim toward colloidal-scale active elements assembling into micromachines that are motile, communicating, shape shifting, and sensing and manipulating their environment. New types of catalysts, fluids with controllable rheology, self-replicating, evolving, and self-healing materials that can store, transform, and deliver energy that conceptually behave like hybrid artificial biological systems. Along with these new materials, we expect new types of phase transitions, new phenomena, and new science.

The ultimate aim of these studies is to find the organizational principles of driven dynamical systems, and we have made some headway. We now know that random collisions under shear strain can lead to exploration of new configurations until the particles no longer collide, they stop, creating an organized absorbing state (Corte et al., 2008). Likewise, topological and symmetry constraints lead to new forms of organization and new types of excitations (Kane and Lubensky, 2014; Bertoldi et al., 2017). However, many of the basic problems remain. Thermodynamic systems tend toward equilibrium. In contrast, driven dynamical systems can tend toward steady state, time periodic, or chaotic constantly evolving states. As the number of degrees of freedom increases and the forces become more complex and different, it becomes harder to avoid chaotic behavior. Familiar properties like entropy and ergodicity become harder to define and measure and their utility in describing phenomena is questionable. Machine learning may prove useful in unraveling the phenomena and even in directing the relevant experiments. There will always be new discoveries as new fuels and materials are discovered. Nonetheless, progress has been made in rational design including spatiotemporal control. Advances are needed in the measurement of active forces and stresses. We would, for instance, like to predict rheology from measurements of the microscopic interactions. New imaging probes and modern microscopy coupled with high-speed cameras can provide an enormous amount of data. Coupled with the data produced by evermore-sophisticated simulations, there is the need for different types of data analysis and the incorporation of artificial intelligence perhaps embedded in both simulation and experiments.

#### **2.1.4 Microgravity**

Most of the active systems we have studied so far are microscopic, colloidal, or suspensions of particles in fluids. They are difficult to density match and, in a terrestrial environment, they sediment, which restricts the experiments to two dimensions. Opening the 3D world with microgravity allows a much greater variety of phenomena, types of organization, and different materials and processing. Further, in terrestrial gravity the driving temperature, concentration, and magnetic (see Figure 1), electric, and light fields create density gradients and produce flows that interfere with the basic phenomena we wish to study. Microgravity allows us to isolate the fundamental interactions and dissect the phenomena. Further, it may be possible to take the particles out of the suspending fluids, as in dusty plasmas (Shukla and Mamun, 2015), and enter a new world where inertia plays an important role and the systems are no longer overdamped. Material processing, such as additive manufacturing, relies heavily on gravity in Earth-bound implementation. In microgravity, new formulations of pastes and slurries are possible. Since future space missions will require fabrication of replacement and spare parts, techniques such as additive manufacturing will be needed. Better understanding of material processing will result from studies both with and without gravity.

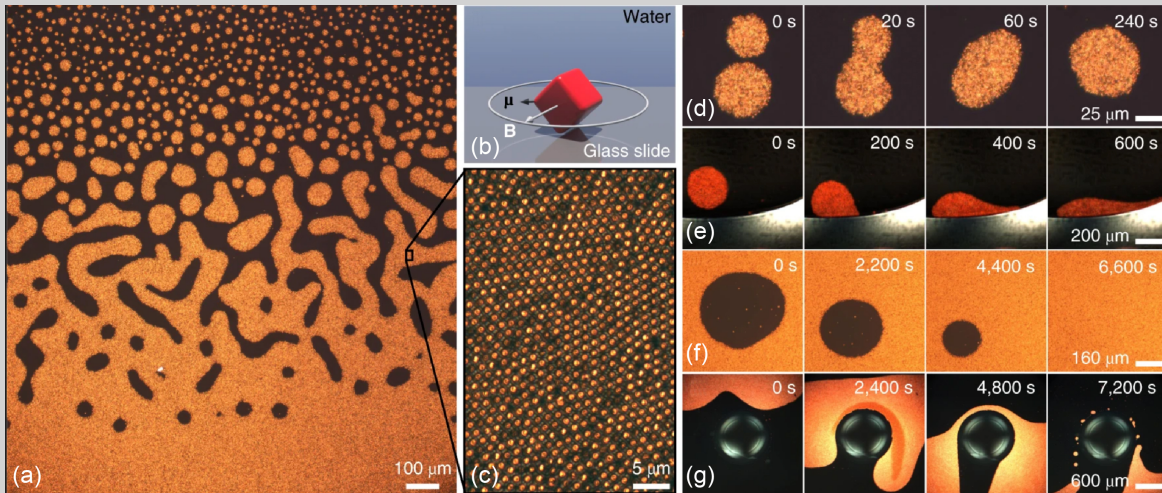


Figure 1.—Chiral fluid of spinning colloidal magnets. (a) An optical micrograph of colloidal magnets in bulk, after few minutes of spinning. (b) Schematic diagram of one colloidal particle. The  $\sim 1.6 \mu\text{m}$  haematite colloidal cubes have permanent magnetic moment ( $\mu$ , black arrow). They are suspended in water, sedimented onto glass slide, and spun by rotating magnetic field ( $B$ , white arrow tracing the white circle). (c) An optical micrograph of colloidal magnets in bulk at increased magnification. Particles attract and form cohesive material with an apparent surface tension that over timescales from minutes to hours, behaves like fluid: (d) clusters coalesce and (e) spread like liquid droplets when sedimented against hard wall; (f) void bubbles collapse; and (g) when driven past an obstacle, fluid flows around it, thinning and eventually revealing an instability to droplet formation. All images were taken through crossed polarizers. Adapted from Soni et al., 2019.

## 2.2 Instrumentation

### 2.2.1 Resource Limitations in Microgravity

Space research is different. There are limited resources, room, power, storage, time, and manpower, and the number of samples and instruments that can be hardened and packaged for delivery even to low Earth orbit is also limited. While microgravity is required for understanding and controlling our experiments on equilibrium and active driven systems, it is anathema for many of the procedures, mixing, transfer, and separations that are routine in terrestrial laboratories. Although microgravity research has provided many exciting discoveries, each one opens new questions and researchers always want to tweak the experiment and try another sample. It is no longer reasonable to do experiments one at a time. The answer is miniaturization. Fortunately, soft matter science often deals with microscopic, micron-scale samples. A nanoliter droplet can contain  $\sim 10^6$  active particles, easily enough for most experiments. A microscope can capture full high-resolution images of this 100- by 100- by 100- $\mu\text{m}$  region. Many of the basic instruments needed for space research have been developed. What is needed is automation, large-quantity sample handling and exchange, and the ability to change sample properties continuously. Flow and external field control can serve a dual role in driving and activating the samples and changing their properties. Commensurate with this increase in samples is the need for faster and smarter data acquisition and system control, which would help to reduce the astronaut's heavy workload.

### 2.2.2 New Technologies

Following on advances in microelectronics and nanoelectronics and photolithography in the late 20th century and the development of soft lithography in the early 21st century, microfluidics and then droplet microfluidics took off (Teh, 2008). The technology developed mostly for the medical and defense industries for high-throughput screening and chemical (especially deoxyribonucleic acid (DNA)) analysis



became an increasingly important technology for soft matter, which studies matter at similar length scales. Thousands of different droplet samples can now be studied rapidly and microscopically using size, fluorescence, particle tracking, osmotic pressure, and a host of other probes. Techniques have been developed for in-droplet mixing and adding and subtracting reagents as illustrated in Figure 2 (Agresti et al., 2010). Biological, chemical, and physical experiments have been performed. More conventional microfluidics also allow exchange of particles and continuous control of chemical environments. The ultrahigh throughput of these experiments will produce large amounts of data, and new data analytic techniques will be required to analyze them fully. Such methods have permeated other bioinformatic fields and will also be applicable here. The 21st century has also seen the development of new optical and acoustic techniques such as time reversal of waves that can focus waves even in turbid media for both observation and manipulation of particles (Lerosey et al., 2004; Fink, 1992). Similarly, video projection and holographic microscopy allow imaging and manipulation (Lee et al., 2007). In addition, high-speed cameras provide unprecedented temporal resolution.

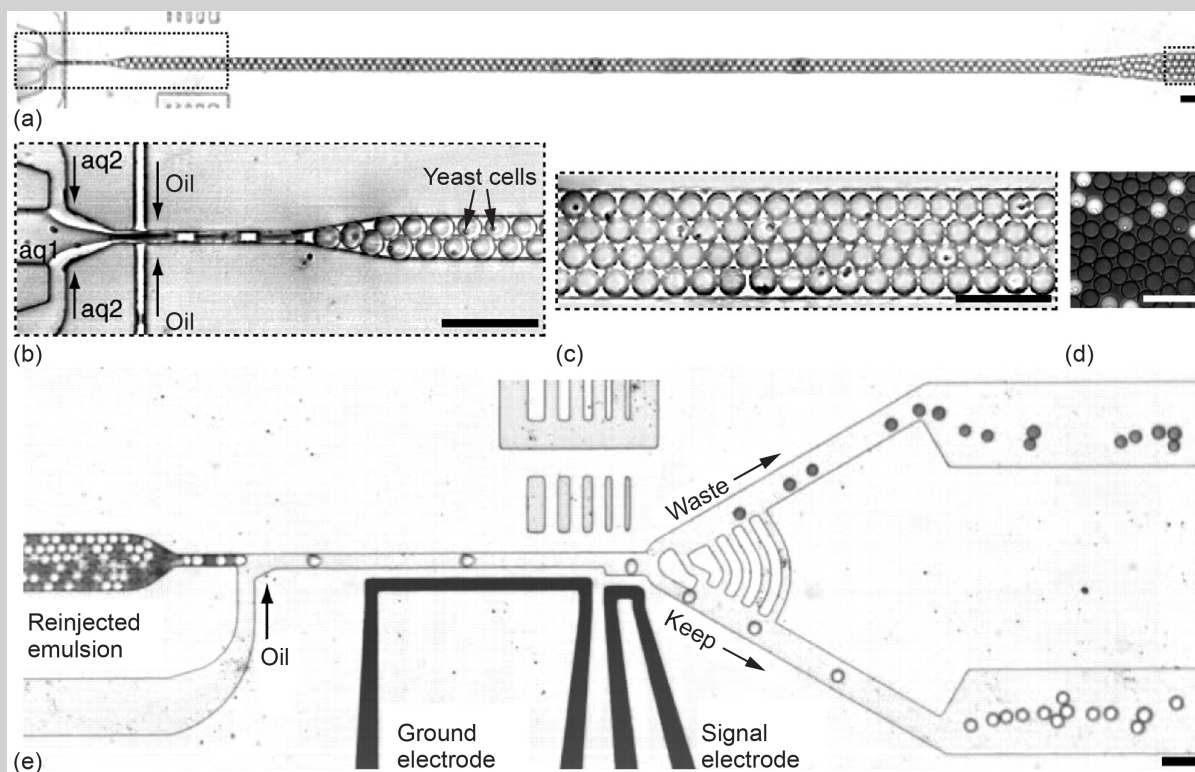


Figure 2.—Modules of ultrahigh-throughput microfluidic screening platform. (a) Low-magnification image of entire drop-making device. (b) Suspension of yeast cells displaying horseradish peroxidase (HRP) on their surface (aq1) is combined with second aqueous stream containing fluorogenic substrate Amplex Ultrared AUR (aq2). Yeast cells are at concentration of  $1 \times 10^8$  cells per mL, which gives an average of 0.3 cells per 6-pL drop after being diluted by half by substrate stream. Aqueous drops are formed at flow-focusing junction (Anna, Bontoux, and Stone, 2003) in fluorocarbon oil at rate of 2 kHz, and number of cells per drop follows Poisson distribution: ~22 percent have single cell. (c) Drops flow out of this device into tube that acts as an incubation line where they incubate for ~5 min. We use fluorosurfactant to prevent coalescence and to give drops a biocompatible interface. (d) Single layer of drops after incubation showing fluorescence developing from active HRP displayed on surface of cells. (e) From delay line, drops flow as solid plug to junction where oil is added to separate drops. To demonstrate the sorting process visually, we sorted an emulsion containing light and dark drops; light drops contain 1 mM fluorescein, and dark contain 1 percent bromophenol blue. Fluorescence levels are detected as drops pass laser focused on channel at gap between two electrodes. When sorting is on, the light drops, which are brighter than threshold level, are sorted by dielectrophoresis into bottom channel. (Scale bar, 80  $\mu\text{m}$ .) Adapted from Agresti et al., 2010.

### 2.2.3 Prospects

From the ideas of instrumentation for studying and manipulating active microsamples came suggestions that similar methods could be used in fabrication and studies of more complex systems. Regarding active units as voxels, elements for almost any material or device can be made on demand using additive manufacturing or self-assembly. With voxels programmable either internally or externally, a wide variety of machines or materials could be produced and then taken apart and recycled for other use. Recycling will certainly play a large role in future space exploration and is important for both active and inactive components. Processed or self-assembled units could produce metafood from plants or recycled organic matter. Designed assemblies of voxels with different properties and activities could make artificial tissues, organisms, and responsive materials that, for example, become stiffer where needed when stressed. A design scheme with submicron particles self-assembled on a fine scale into an active cluster and then 3D printed on a macroscale will allow heterogeneous structured materials as, for example, nature does in bone.

The next generation of soft matter experiments, especially those involving nonequilibrium phenomena, will involve new ways of controlling a particle's position, density, and activity, new ways to measure linear and nonlinear dynamic response, and new types of organization. Much of the research is readily done with advanced optical microscopy. Microrheology (Mason, Gang, and Weitz, 1996; Gittes et al., 1997) now let us probe the viscous and elastic properties of microscopic samples. Particularly important will be new probes that measure forces and stress, similar to those used in microbiology where different types of chemical and electrical membrane activity are observed with optical probes (see Wikipedia article on Biosensors). Many active systems are driven by phoretic effects, (motion in self or externally generated field gradients) as, for example, thermophoretic, electrophoretic, or diffusiphoretic effects. Future experiments will combine phoresis with flows generated by microfluidic devices and particles rotating and spinning in magnetic and electric fields. Instruments will have to accommodate a variety of fields and flow control with fast high-resolution imaging and data processing with integrated feedback perhaps overseen by machine learning. Especially for microgravity research, miniaturization and high-throughput instrumentation will be required to optimize output under time and space constraints.

### 2.2.4 Microgravity

The advantages of microgravity research include, particularly, the exploration of three dimensions rather than two and the absence of uncontrolled convective flows. The price is the reduced time and space for experiments. These can be addressed by miniaturization of the samples and development of instrumentation that can provide high-throughput data acquisition and feedback control of the quiescent and dynamic samples. Additive manufacturing will also play a substantial role in several aspects of microgravity research and later in space exploration. Experiments on the pastes, slurries, and new rheological materials that are processed in 3D printing will advance additive manufacturing both on Earth and extraterrestrially where the processing materials and protocols will be different. The technology developed will be important for preparing and processing experimental samples. Later recycling of materials to make devices, instrumentation, repair and replacement parts, and even prosthetics, will allow a sustainable environment for exploration. New methods for fertilizing and growing plants, as well as new methods of making plant-based foods, such as meats, will have to be adapted to microgravity. These could help provide a sustainable source of nutrition for very long spaceflights.

## 2.3 Suspensions, Foams, Emulsions, Colloids, and Granular Materials

### 2.3.1 An Explosion of New Building Blocks

Particles, droplets, and bubbles with and without surrounding fluids fill the world we experience every day and their study has engaged humanity from antiquity. So has their utilization. The first processed colloids were inks and paints with particles stabilized with surfactants such as the gum Arabic used in Egypt. Presently, they are industrially important from concretes, ceramics, and food to biotechnologies and medicine. In soft matter, they also serve as model systems for understanding the organization of matter from the atomic to the cosmological scale. The fact that they are observable in real space and real time has led to fundamental insights into transitions such as crystal-liquid melting and the formation of glasses (Pusey and van Megen, 1986). The basic question to be answered is how the elementary building blocks and their interactions and processing lead to bulk macroscopic properties. These are real-world problems involving hydrodynamic interactions, gravity, friction, charging, and hysteretic effects in both self-assembly and manufacturing.

Throughout the 20th century, the general field of colloidal science dealt with spherical particles and a few elementary forces, electromagnetism, Van der Waals attraction, excluded volume, and depletion interactions leading, for example, to crystals with simple symmetries. The 21st century has already seen a revolution in the types of particles available and a host of new interactions. There are now a plethora of shapes: ellipsoids, cubes, Janus and patchy particles, and even shape shifting particles (Hong, Jiang, and Granick, 2006; Youssef et al., 2016).

All of these can now be functionalized to have specific interactions that are programmable for complex self-assembly. Lock and key particles only fit together as their name implies; DNA and, more recently, protein-coated particles and droplets bind only to their complementary forming flexible or rigid chains; designed finite clusters and crystals as complex as diamond (see Figure 3) and clathrates can be assembled (Lin et al., 2017; He et al., 2020). Further we can now program sequential self-assembly of arbitrarily designed structures controlling even the handedness of their chirality (Zion et al., 2017). Other sections of this report elucidate how these particles and constructs can be activated. Advances in simulations and machine learning have enabled the use of these experimental tools in assembly of new materials and constructs. They have also helped elucidate the role of entropy in structure determination (van Anders et al., 2014).

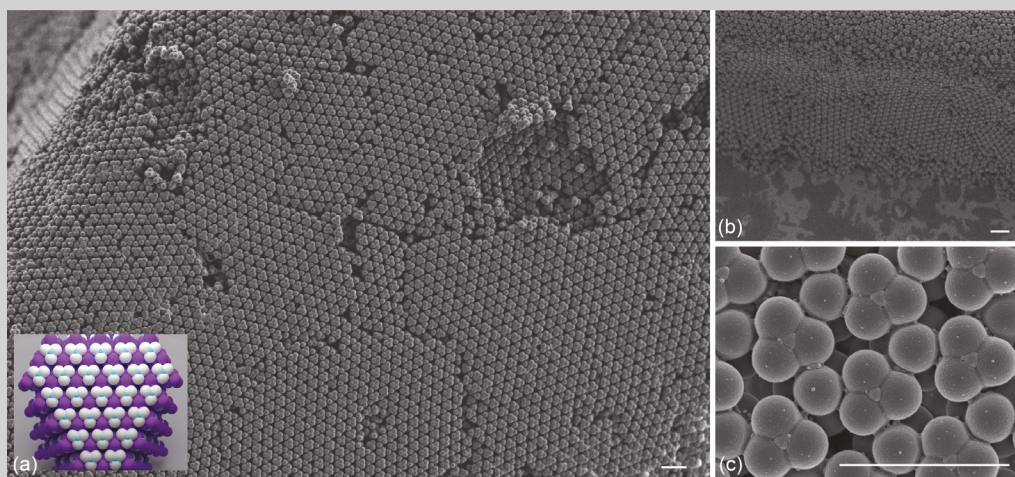


Figure 3.—Flexible probe being inserted into photoelastic granular material, performed in three different gravitational conditions (via parabolic flights), illustrating qualitative changes in behavior under reduced gravity. Bright particles are those experiencing higher local forces. (a) Microgravity. (b) Lunar gravity. (c) Martian gravity. Adapted from Featherstone et al., 2020.

The past several decades have seen the cross-fertilization of these different subfields and materials in soft matter. Granular materials are dominated by gravity and friction, which have found utility in novel mechanical and robotic artificial hand grippers (Brown et al., 2010). Recently, it has been realized that frictional effects are also important in colloidal suspensions as well as being involved, for example, in shear thickening (Lin et al., 2015). Granularlike colloidal behavior is also involved in the arrest of phase separation in the formation of bicontinuous gels (Cates et al., 2004), a labyrinthine form of a Pickering emulsion. Colloids anchor nematogens in liquid crystals (LCs) and direct topological defects and even the formation of knots. Colloids can also be suspended in plasmas, where inertia is important, leading to new nonequilibrium phenomena akin to hydrodynamic turbulence (Gogia, Yu, and Burton, 2020).

### 2.3.2 Prospects

While we have made great advances in understanding and controlling the self-assembly of particulate systems, the basic problem of going from microscopics to bulk properties is yet unsolved both for atomic and molecular and for colloidal systems. However, the processing of these systems in both equilibrium and nonequilibrium has, and will, lead to useful materials. For example, we have learned how to make equilibrium photonic crystals (He et al., 2020) and are on the track of isotropic hyperuniform photonic bandgap materials by nonequilibrium shear (Florescu, Torquato, and Steinhardt, 2009). Machine learning should point the way toward such a theory and statistical mechanics of building blocks with fixed or stimulus-responsive properties. A target might be a slurry with a variety of properties that can be tuned on demand. Recently, we have self-healing materials (Cordier et al., 2008). In situ resource utilization is especially important for space exploration and will inform terrestrial sustainability. Recycling, purification, and concentration systems rely on liquid interfaces, bubbles, and foam emulsions and dispersions. Advances are needed since present recycling efficiencies are on the order of 20 to 30 percent.

### 2.3.3 Microgravity

There have been more than two decades of development by NASA and ESA on instrumentation for colloidal research in microgravity, especially advanced microscopy. As noted in other sections, the problems are in the number of samples that can be run and in the crew time. We, therefore, need miniaturization and automation. Rheological measurements are of particular interest for this class of materials some of which are accessible by microrheology (Mason, Gang, and Weitz, 1996).

Gravity is highly relevant for the dynamics of granular materials (Guyon, Delenne, and Radjai, 2020), where it is the dominant interaction in many terrestrial experiments, particularly as regards industrial handling and processing. In foams, gravity-driven drainage is an important problem (Weaire and Hutzler, 2001) and in suspensions, pastes, and slurries, particle sedimentation influences, and often obscures, the interesting science and affects the processing protocols (Guazzelli and Morris, 2012). Therefore, microgravity is a particularly useful knob for testing the relative importance of various mechanisms: the ability to provide different degrees of gravity is a unique tool for fundamental research (see Figure 4 for example). Tunable processes such as self-assembly and manufacturing will be different in microgravity, potentially leading to new applications.

Space exploration provides some additional challenges and opportunities for particulate research (NASA, 2020). In low-humidity environments, charging effects from both unblocked radiation and tribocharging of vibrating grains can lead to hazardous circumstances for the missions themselves. An improved understanding of granular material under such conditions will provide important fundamental knowledge needed to explore fully planetary and other extraterrestrial bodies, particularly those with powdery surface regolith, and in particulate rings. Grains suspended in a plasma (dusty plasmas (Shukla and Mamun, 2015)) have been studied for more than a decade in microgravity conditions, so there is

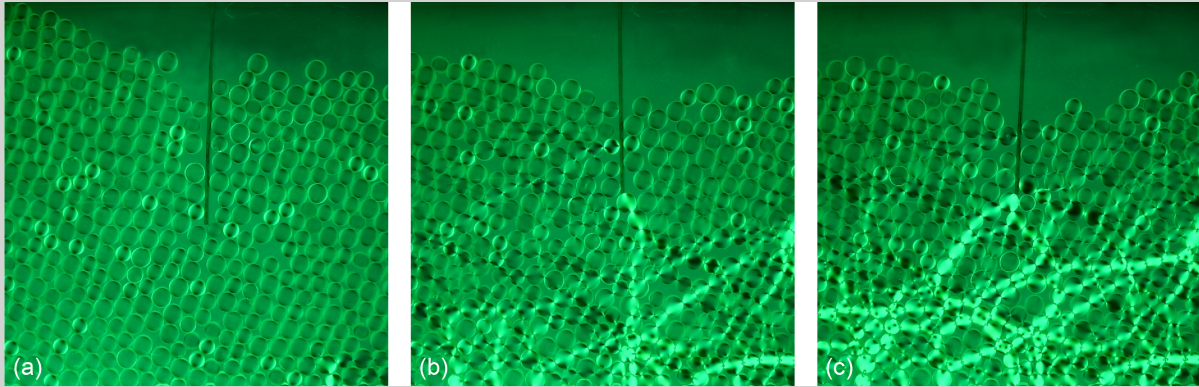


Figure 4.—Patchy, triangular, deoxyribonucleic acid (DNA) functionalized particles and crystallization of cubic diamond colloidal crystals. (a) Scanning electron microscope (SEM) images of 111 plane of colloidal diamond crystals. Crystals are about 40  $\mu\text{m}$  across, with grain boundaries and point defects. Inset, computer-generated image showing 111 plane of colloidal diamond crystal for  $d_{CC}/(2a) = 0.74$ , consistent with SEM image. (b) Side view of crystal edge. Thickness of crystal is about 10 to 20 particles. (c) Magnified SEM image of 111 plane showing interlocking of particles, as designed. For particles shown in (a) and (b),  $d_{CC}/(2a) = 0.73$  and  $b/a = 1.20$ ; in (c)  $d_{CC}/(2a) = 0.75$  and  $b/a = 1.19$ . In (a) to (c), scale bars are 5  $\mu\text{m}$ . Adapted from He et al., 2020.

potential for cross-fertilization of ideas to tackle engineering challenges. Another consideration is the importance of conserving resources in spaceflight and extraterrestrial habitation, including those obtained by in situ resource production. Recycling efforts would certainly need to exceed what is presently the norm on Earth, and the mining or reprocessing of materials would often entail handling many types of particular materials of this class. The techniques developed for space exploration will feed back to improve sustainability and life on Earth.

## 2.4 Packings, Simulations, and Big Data

### 2.4.1 Machine Learning and Big Data

Many branches of science depend on handling large sets of data. This is true in high-energy experiments, in weather prediction (and on a longer scale climate prediction), for cosmological simulations of the universe's expansion, for studies of proteins in biology and, of course, in soft matter. It is the latter subject that we concentrate on here.

Simulations of soft matter systems produce prodigious amounts of data that include, but are not limited to, positions and velocities of all the particles in a large system. Large systems are crucial for any study that hopes to make contact with the underlying statistical mechanics of the system at hand. In the case of simulations of glasses, where in addition to there being many particles, the dynamics are also very sluggish, not only are large numbers of particles required but very long timescales are needed to sample the dynamics as the system slows down (Debenedetti and Stillinger, 2001; Berthier et al., 2019). Even if the dynamics are not an issue, any search for hidden structure inside of disordered systems or particle packings requires massive resources to store and process the data.

Many soft matter experiments are also done by acquiring and analyzing a large number of images. Some of these are ordinary still photographs but others are videos of events such as those that occur in fluids or in active-matter experiments. When living matter is involved, even more data is often required.

While this reliance on computer-assisted data analysis is not a recent development, advances in modern data science and machine learning provide new opportunities for gaining insight into natural phenomena, whether based on simulations or experimental data. Indeed, soft matter scientists and engineers bring something to machine learning because the problems in soft matter are often subtle enough that no easy answer is available from more ordinary methods of analysis yet there is sufficient data so that the computer can learn to make the appropriate distinctions. Thus, there are cases where, by resorting to machine-learning techniques, new science has been discovered (Schoenholz et al., 2017).

One issue that naturally arises is whether there are universalities in how machine-learning methods work. Do those techniques succeed by coarse graining the system or do they work by some other, not yet understood mechanism? Are there hierarchies involved and is the behavior emergent? These questions bring us to the issue of what is the physics involved in machine learning itself and can it be understood in terms of known statistical mechanics concepts. Soft matter (in particular, the physics of glasses and jammed systems) is an excellent starting point to learn about how these various concepts are related (Geiger et al., 2019). Can insights from the study of glasses and jamming feed back into the general understanding of data science?

A common approach in statistical mechanics is to coarse grain a system into larger and larger entities in order to isolate the emergent behavior. Machine learning can identify (i.e., learn) the relevant aspects of the system and apply them to nonequilibrium models. It can help achieve good techniques for coarse graining.

One particularly intriguing possibility is to imagine an analog to machine learning that takes place, not in the software of a computer, but rather in the elastic behavior of a mechanical system. This possibility is suggested by the recent ability of being able to tune network-based materials by training them to perform a desired behavior (Pashine et al., 2019; Hexner, Liu, and Nagel, 2018). The idea of training is in some context an old one, but recent advances have suggested that training can be used as a much broader paradigm than has been realized previously for how to create materials with a large variety of possible behaviors. There should be a two-way street between machine learning and the statistical mechanics and the theory of elasticity. That is, lessons learned from manipulating materials in the laboratory should not only borrow from the techniques developed in data science but also be able to be transferred to analyses being made inside a computer that is trying to learn how to distinguish and classify different inputs.

The studies that are being currently done to see if materials can be trained to have desired outcomes can also be generalized to the algorithms themselves that are being used. Can we learn to adapt the algorithm to achieve optimal performance so that it is possible to get past bottlenecks in computing by letting the algorithm itself learn what is hindering the progress? In addition, there is also the possibility of creating quantum algorithms for linear programming of relevance to soft matter and glassy systems (Bapst et al., 2013).

#### **2.4.2 Prospects for Jamming and Packing**

Sluggish dynamics is at the heart of glassy phenomena and has been an integral part of much soft matter research. Importantly, there have been some recent advances in the area of glass physics that have reinvigorated this field. After decades of effort, there has at last been spectacular developments in at least two areas. It is not only exciting in itself, but it is also relevant because there are spinoffs to the glass problem that can have great impact on a wide variety of scientific endeavors.

One astounding success in the area of glassy simulations is based on a new implementation of the swap algorithm (Berthier et al., 2016; Ninarello, Berthier, and Coslovich, 2017). In that approach, an added degree of freedom is included in the Monte Carlo code so that two particles with different sizes can exchange positions if energetically favorable. This is a nonlocal move that would not be allowed with more conventional simulation algorithms. With swap Monte Carlo, the simulations are able to go beyond what even experiments can do to allow the particles to equilibrate at low temperature. This phenomenal success opens the question of whether this or a similar algorithm, can be adapted elsewhere for other types of problems. In particular, it allows access to far-from-equilibrium behavior where thermal relaxation is no longer possible. Out-of-equilibrium packings can be very different from equilibrium ones. Thus, techniques similar to swap would be very useful for accelerating nonequilibrium simulations if that turns out to be possible. Machine learning might be able to teach us how this might be possible.

Glasses in the infinite-dimensional limit are also becoming well understood theoretically (Parisi and Zamponi, 2010; Berthier et al., 2019). What are the hallmarks of the infinite-dimensional system that appear in the low dimensions? It is known that there are big differences in low dimensions that appear universal but do not emerge out of the infinite-dimensional solution. One issue that is currently being investigated is whether or not these low-dimensional effects are perturbative. If not, where do they come from? These studies have demonstrated strong connections to network theory and have opened up new approaches to understanding the two-level localized excitations that are responsible for the low-temperature properties of disordered solids (Khomenko et al., 2020).

In a frictionless jammed system, there is an exceedingly rugged energy landscape of allowed packings. At high density, the configuration space gets very tortuous. In part, the glass problem is connected to how systems explore that landscape. There are several modes of exploration including varying the temperature or shearing the sample. These are the two most conventional ways that the exploration has been done. Recently, swap Monte Carlo has provided another way. A fourth way is to prune bonds from the network; the network goes from one ground state to another depending on what bonds were pruned (Goodrich, Liu, and Nagel, 2015). In the context of training the network to adopt some particular function, it needs to be determined which variables one can train on and which ones cannot be used. Can these variables be categorized into different types? By mapping the glass onto a network, this makes a bridge to network science. This suggests that the geometric part of packing might be universal. Protocols for pruning or training greatly change how that network evolves. One question that emerges from this study of the energy landscape is whether there are other, different, ways of exploring the landscape schematically shown in Figure 5. Could we use data-science methods, such as machine learning, to map the landscape? An additional frontier is to understand which of these answers carry over to experimental systems in which friction and/or adhesion are present: is the energy landscape still a valid framework when there is dissipation?

What has also become apparent is that active systems can also develop slow dynamics. Systems sheared to failure undergo a transition that is reminiscent of the behavior of dense active matter. This provides a foundation for studying these systems in parallel to see how they are similar and how they are different (as indicated in Figure 5). These issues can be broadened further to ask how they are similar to systems driven in other ways. Are there formal similarities between different dynamics, for example, shear and active forcing (Nandi and Gov, 2018; Morse et al., 2020)? In addition, the modes of an active system look qualitatively very similar to those in the bulk of a jammed system (Henkes, Fily, and Marchetti, 2011; Bi et al., 2016; Henkes et al., 2020).

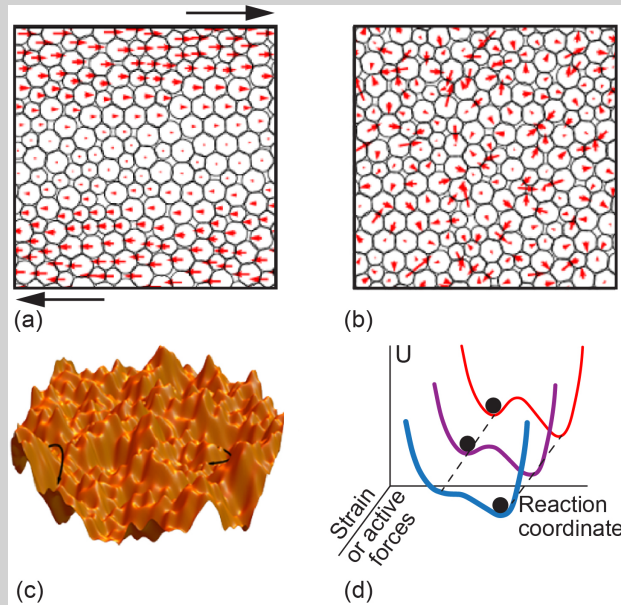


Figure 5.—Traversing complex potential energy landscape in soft matter packings. Two different methods for traversing potential energy landscape in a packing of particles: (a) applied shear strain and (b) active forcing. (c) Schematic of complex potential energy landscape as function of two coordinates; in soft matter systems, such landscapes are at least  $Nd$ -dimensional, where  $N$  is number of particles and  $d$  is dimension. In swap Monte Carlo and similar algorithms, where radius of particles can vary, there are  $N(d+1)$  coordinates. (d) Schematic of potential energy  $U$  as function of an applied displacement, such as shear (a) or active forces (b), and reaction coordinate along which system encounters a saddle. This is how systems explore new minima; applied strain and active displacement fields generate very similar dynamics. Tuning and aging are other optimized methods for traversing configuration space in networks. (a), (b), and (d) are adapted from Morse et al., 2020. (c) is adapted from Biroli, Bunin, and Cammarota, 2018.

### 2.4.3 Biological Matter and Food Science

The science of glasses bears an obvious relevance to the problem of protein folding as well as other biological functions. Indeed, jamming, packing, and granular physics appear in tissue morphogenesis and in the cytoskeleton (Merkel and Manning, 2017). Biological materials have a strong connection to metamaterials, and indeed, cellularized tissues and fiber networks rigidify (Sharma et al., 2016; Merkel et al., 2019; Yan and Bi, 2019) in a way that shares many similarities with metamaterials such as origami (Chen and Santangelo, 2018) shown in Figure 6. Similarly, allostery (long-range control between local sites in proteins) can be found in mechanical networks (Rocks et al., 2017; Rocks et al., 2019; Yan et al., 2017). It is an inherent property of dense, disordered connected systems. Transitions to rigid structures in food science could be important although it is not very clear yet if that rigidity transition is related to jamming in dense packing of particles.



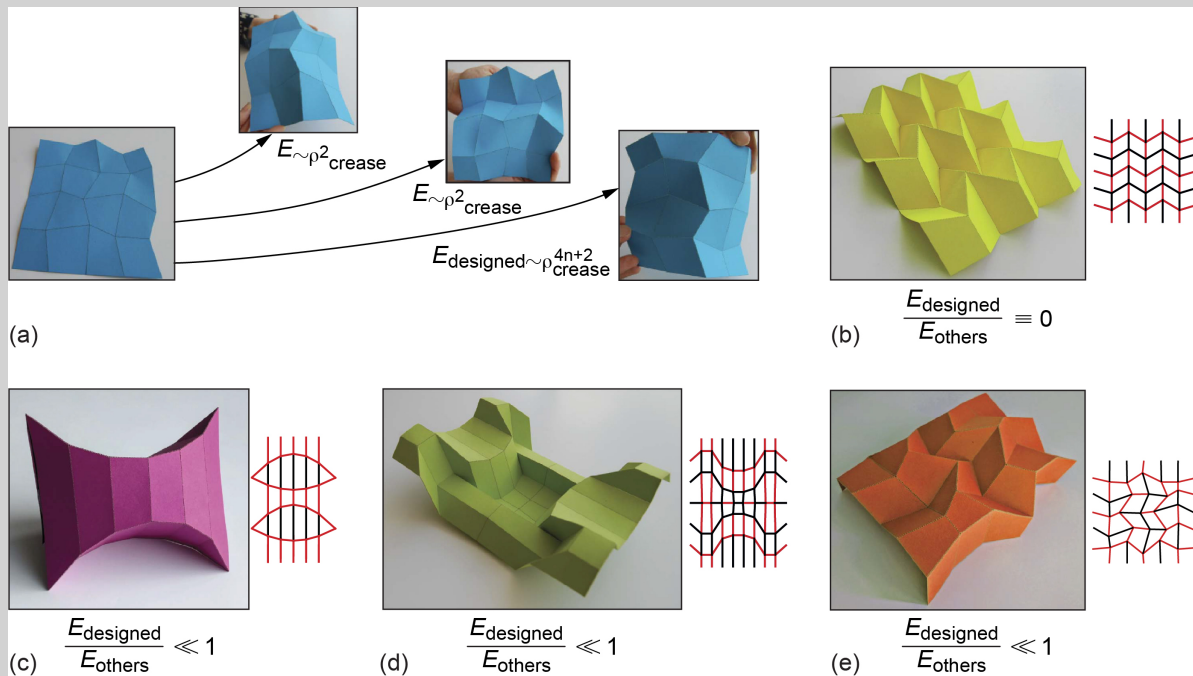


Figure 6.—Origami patterns. (a) Forces applied to “self-folding” sheet will preferentially actuate one pathway designed to have significantly less face bending than other two pathways shown (that is, designed to have  $E_{\text{designed}}/E_{\text{others}} \ll 1$ ). (b) Celebrated Miura-Ori pattern is special highly symmetric pattern with  $E_{\text{designed}}/E_{\text{others}} \equiv 0$ . (c) to (e) Larger space of experimentally relevant crease patterns obtained by going beyond rigidly foldable symmetric patterns. Folding energy scale of such patterns can be made as small as needed in systematic manner. Patterns in (c) to (e) are geometrically distinct from traditionally studied limits (Kawasaki vertices and Miura-Ori Mountain-Valley choice). Adapted from Pinson et al., 2017.

#### 2.4.4 Microgravity

There are some special advantages of microgravity for the research on particle packings. In terrestrial laboratory studies of jamming, gravity will always play a dominant role. Microgravity would allow a way to circumvent the gravitational interactions so that other aspects of jamming can be studied carefully in the laboratory. In studies of human crowds and other active matter systems attracted to a point of interest, it is observed that there are strong density and pressure gradients from the edge of a crowd to a point of interest (Bottinelli, Sumpter, and Silverberg, 2016). Very little seems to be known about such systems. Microgravity would be an ideal place to study the effects of pressure gradients in jammed systems.

There are specific topics that have relevance to NASA. For example, the NASA landers have had trouble digging into the landscape of Mars. Why is this the case? Another topic is that aggregation of dust particles to form a planetesimal is difficult when there are no strong gravitational interactions; the key interactions are now intermolecular and electrostatic. Thus, the study of aggregation in microgravity would have a strong basic science motivation.

Studies of packings, even in microgravity, will produce a high volume of image and video data so machine learning will be essential for its analysis.

### 2.5 Mechanical Metamaterials and Topological Soft Matter: Allosterity and Auxetics

#### 2.5.1 Metamaterials: Inverse Design and Additive Manufacturing

One area that has recently drawn the attention of soft matter scientists and engineers is the possibility of designing so called metamaterials that have their functions governed, not by the specific substance out of which the material is constructed, but rather by its microstructure. Illustrative examples of

metamaterials are origami folded from a two-dimensional (2D) sheet or laser-cut out of rubber as shown in Figure 6. In such origami, the pattern of folds in the sheet governs how the sheet can fold into a desired 3D object (Bertoldi et al., 2017).

A vibrant research area is to map the possible reach of metamaterials functionalities. In the future, one could input into a computer (or experimental protocol) the desired property and have as an output the appropriate microstructure that would produce it. Such metamaterials could then be produced using additive manufacturing techniques. A natural question emerges: is there a way to create bulk quantities of folded or extremely deformable metamaterials without building them one piece at a time?

Another challenge is whether (meta)materials can be designed to have multiple functions encoded in their structure. This requires understanding what platforms for the underlying material would be amenable to this form of manufacture. For example, what material is best for 3D printing? It also requires understanding what types of functions can be programmed into a metamaterial. This problem would benefit from machine-learning techniques. Basic questions include (i) to what extent can multipotent metamaterials be designed so that they are capable of having multiple functions accessible upon deployment and (ii) is there a limit to how many functions can be encoded into a given structure.

More generally, in creating useful metamaterials, one typically starts with a goal and then tries to guess the structure able to accomplish it. What kinds of protocols can be used to solve this challenging inverse problem systematically? Making use of data as a learning algorithm is an enticing route to endow a material with a good protocol for generating desired functions.

### **2.5.2 Adaptable Materials**

One tantalizing idea is that designed materials could be sufficiently adaptable to produce a desired outcome via training, that is, materials learning. In the area of network-based materials, pruning of specific bonds has been shown to be a viable strategy for imparting function (Goodrich, Liu, and Nagel, 2015). How can this be generalized? What other ways can we use to tune the properties of the resulting material? Certainly, one can think of adding other forms of complexity through structure, shape, interactions, and possibly activity that allow for an even broader range of functionalities.

If one departs from network materials, where all the bonds are specified at the outset, one arrives at a much more difficult problem. It is a big challenge to develop control over particulate systems where the particles are free to rearrange under an applied force. It is not clear, in that case, whether it is possible to even train functions in the same way as for the simpler case of networks. However, it is conceivable that the individual building blocks can have information built into them. For example, as in DNA-coated colloids, one can design specific interactions between particles of given types. One can generalize such adaptability to include more complicated information at each site.

One can also think of adaptable materials as a platform for some form of computation and memory formation (Keim et al., 2019). Thus, materials learn to distinguish between different classes of inputs imposed from the outside. This is machine learning where the machine is not a computer, but the physical material itself (Stern et al., 2020).

### **2.5.3 Active and Self-Sensing Bioinspired Metamaterials**

The microscopic constituents of a material or metamaterial, be it particles, bonds, or hinges, can be activated, that is, they can have their own source of energy as it occurs, for example, in biological tissues and filaments with motors. In addition, synthetic materials that use biological components give rise to novel collective phenomena and responses (Sanchez, 2012). Self-sensing metamaterials built out of robotic components (Y. Chen et al., 2020), like the active metabeam in Figure 7, displays feedback between local deformations and activity leading to new “odd” elastic moduli associated with distributed

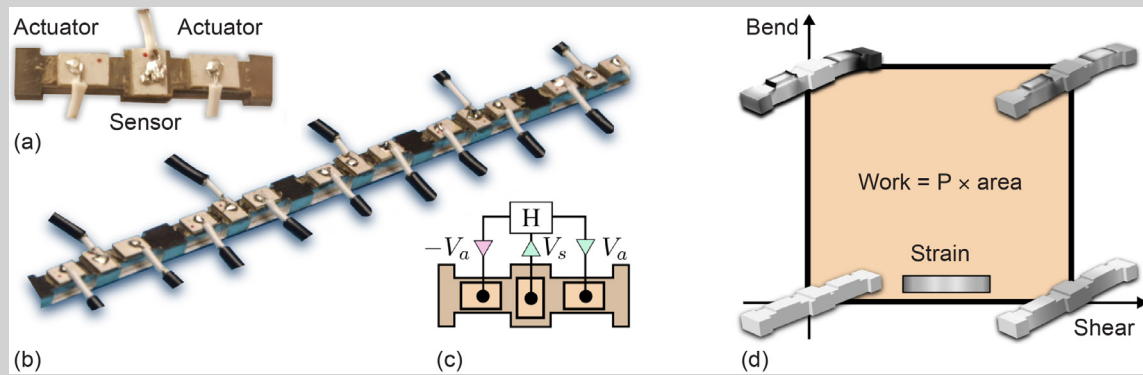


Figure 7.—Self-sensing metabeam utilizing minimal onboard computation. (a) Single unit. (b) Multiple units connected together. (c) Each unit cell consists of steel host beam equipped with three piezoelectric patches: central patch senses deformation and sends an electrical signal to outer patches, which actuate forces. (d) Feed-forward electromechanical results in an emergent odd elastic modulus  $P$  that cannot be derived from potential energy. Odd modulus is proportionality constant between area enclosed in deformation space and work extracted during mechanical cycles involving shear and bend. Figure adapted from Y. Chen et al., 2020.

energy cycles (Scheibner et al., 2020) Similarly, grains and colloids with motors can flow and transmit sound in unprecedented ways and exhibit exotic hydrodynamics (Marchetti et al., 2013) and phase transitions (Fruchart et al., 2020).

If we think of making active materials useful for biological implants, we must face the challenge of using biocompatible materials in additive manufacturing (e.g., 3D printing). Biophysical systems are inherently active matter. Molecular folding of proteins, tissues, and chromatin are key examples. A 20-year goal could be to create adenosine-triphosphate- (ATP-) driven molecular systems with spatiotemporally controlled activity. Such systems would involve chemistry-based nanoscale building blocks with controllable activated processes. A further goal would be to formulate topological design principles that protect the desired functionalities against noise and manufacturing imperfections (Shankar et al., 2020).

#### 2.5.4 Integration of Computers and Materials

Information is physical. In his famous lectures on computation (Feynman and Hey, 2000), Feynman illustrates this mantra by means of thought experiments of computing with biological components. Soft biosystems are gradually emerging as viable platform for information processing and computation. However, we still think of materials, computers, and machines as distinct. The materials of the future will no doubt blur these boundary lines. Tasks such as autonomous information processing, microscopic energy transduction, and reconfigurability will need to be incorporated into materials design without recourse to external batteries or computers. There is, in fact, a deep connection between information and thermodynamics. A material platform that processes information autonomously in a cyclic manner must necessarily be active. Even if all sources of dissipation were to be minimized down to theoretical limit, erasing information upon resetting memory would always entail an energy cost (Plenio and Vitelli, 2001). The comprehensive integration between material design and computation required to create autonomous computing materials will entail a deeper theoretical understanding of the interplay between information science, material synthesis, and the physics of active matter.

#### 2.5.5 Microgravity

Extreme soft matter mechanics: microgravity is the realm of the superweak, and potentially of the self-assembly of the superlarge. A famous example is the ISS-based diffusion-limited aggregation of submicron-sized colloidal particles into ultraweak fractal clusters of centimeter dimension (Lu et al., 2008), which would otherwise be crushed by 1g. But now we are pushing the limits of the directed

self-assembly of structures to larger length scales and more complex structures, processes demanding microgravity. It is possible that there are many liquids, including water, that are actually ultrasoft solids, but are pushed beyond their elastic limits everywhere on Earth by the combination of gravity with geothermal dynamism, convection, and thermal activity due to solar heating.

## 2.6 Soft Matter, Bioscience, and Biotechnology

The field of soft matter began with early humans learning to deal with the soft, which apart from air and water, were largely the materials of life: hierarchical assemblies and composites spanning the full range of structural behaviors from liquid to solid, driven and evolved into active information-rich nonequilibrium states and structures by solar power. After millenia, the emergence of modern science was finally able to disentangle the science of soft matter from the science of life, and pursue the physics and chemistry of polymers, gels, emulsions, surfactants, colloids, and LCs as fields of materials research. Consequently, the cross-fertilization between soft matter and biology has been extensive, and now, in a turnaround, moves toward imbuing the broad range of soft materials with more lifelike properties, an emergence largely evolved by the ingenuity of chemists. The building blocks now have properties such as activity; recognition capability; chemical, temperature, or light sensitivity; and the ability to change shape.

One theme that arose in the workshop was why not go one step further and let them “think” and communicate. A basic question then is how to understand, control, and use such a complex dynamical system, so that it does not destroy itself chaotically (Wu et al., 2017). It appears that such questions will drive much of 21st-century soft matter science. While 19th-century science invented equilibrium thermodynamics and statistical mechanics, and the 20th century learned to apply these methods to understand the equilibrium properties of soft matter, the focus of the 21st century is nonequilibrium statistical mechanics, phenomena, and dynamics, ranging from the physics of swimming to understanding the origin of life.

### 2.6.1 Polymers

There is a major difference between synthetic polymers such as polystyrene and polyethylene in solution and biopolymers such as polypeptides and nucleic acids (NAs); the bonds keeping the chains intact are stable for the synthetic ones but only metastable for those that occur in biology (Dobry et al., 1952). This is evident given that there are nonactive enzymes that are able to cleave the peptide or sugar linkages in biopolymers. Polymer theory should be reexamined for this class of metastable, living polymers. In particular, the entanglements of the polymers will be essentially different and needs to be worked out in detail. This has been partially investigated for somewhat similar cylindrical microemulsions. More specifically the rheological behavior, especially in the presence of the cleavage enzymes, should be investigated both in vitro and in vivo. This presents one significant challenge for the field.

The metastability of the biopolymers could be even more significant for the case of intrinsically disordered proteins (IDPs), which are often polyampholytes. IDPs exhibit many biologically relevant instances of protein-protein phase separation. This has been well documented in the last few years (Yongdae and Brangwynne, 2017). These molecules provide a whole set of new degrees of freedom where the polymers could become shorter or longer by breaking as well as by stretching. The field needs to be revisited in detail. The metastability coupled to active substrates, motors, chemical reactions, and other energy-dissipating processes may lead to a rich landscape of phase behaviors from equilibrium (no active elements) to dynamic (in the presence of active elements).

## 2.6.2 Prospects

Hierarchical assembly has been an active area of research for decades (see the Nation Research Council report “Hierarchical Structures in Biology as a Guide for New Materials Technology” (1994) for example). However, multiscale active biological structures such as microtubules, chromatin, or neurofilaments have not yet to our knowledge been achieved in synthetic systems. One of the experimental constraints is that gravitational sedimentation or creaming limits them to nanosize objects. However, in microgravity, one can make much larger entities. Thus, an important challenge with many potential applications is in the direction of self-assembly of large objects where microgravity could play a pivotal role.

Over the last few years, it has become increasingly clear (Smith, Lee, and Perkin, 2016) that the range of validity of Debye-Hückel theory of electrolyte solutions (i.e., salty water) is limited to tens of millimolar concentrations for even one to one salts (e.g., NaCl). This calls for new methods to measure electrostatic screening such as the use of x-ray and neutron scattering (both elastic and inelastic) to probe ion-ion correlations in polar solvents. The theoretical situation also remains murky. While an established field, it is crucial for the control of biomolecular assembly, and merits clarification. For example, the old problem of the ionic-strength dependence of the polyelectrolyte persistence length is very likely associated with this issue.

It is important to emphasize that there are many basic aspects of soft matter equilibrium that have not yet been fully developed. These include Debye screening and electrostatics, for example, where behavior at high ionic strength is a dynamic current research field. Likewise, there are many unanswered questions regarding hierarchical self-assembly, of artificial microtubules, for example. In addition, the physics of protein-protein phase separation, IDPs and multiple structures, and issues having to do with polymer solutions, polyampholytes, and polyelectrolytes need more clarity.

Another very broad challenge has to do with dealing with systems, with or without activity, that are far from equilibrium. In the context of biology, this emphasizes complex (in charge and sequence) polymers and includes understanding motility-induced phase separation (Cates and Tailleur, 2015).

## 2.6.3 Soft Matter and the Origin of Life

The advance of biology in the 19th century fostered an intense scientific interest in understanding the origin of life, which developed in the 20th century with the flowering of biochemistry and cell biology, principally around the notion that the mystery of life’s emergence was a problem in chemistry. Some remarkable insights were gained in this effort, not the least of which was that in the universe and early Earth, there were amino acid, NA, and lipid components, as well as accidental chemical reactions that could produce them from primitive molecular species. As this work progresses in the new century, the understanding of the physical states and collective soft matter organization of prebiotic systems have received increasing attention as key parts of the story.

The emergence of early life, by definition, requires feedback: molecular selection and replication self-organized around some purpose, which in the earliest stages must be the stabilization and protection of active soft matter self-assemblies: “Soft matter” because it permits the coexistence of structure and chemistry, and “active” because the overall process is inherently nonequilibrium. Hierarchy develops as complementary feedback systems begin to interact, and all this evolves in the realm of marginal solubility, where the insoluble molecules are lost, the readily soluble are just part of the solvent, and the action is among molecules that must collectively interact into order to stay in solution. Figure 8 and Figure 9 show this in action for lipids and NA oligomers in which, for the latter, LC columnar ordering strongly promotes NA ligation and thereby the stability of the columnar phase (Fraccia et al., 2015). The grand challenge is to advance the science of templating and molecular replication by soft matter (He et al., 2017) to the state in

which the regime of accidental chemistry can be connected with that in which autocatalysis stabilizes and evolves collective ordering.

### 2.6.4 Microgravity

Several problems related to biological activity such as hierarchical self-assembly, particularly from activity, would profit from making isolated model systems. This recalls the question of MIPS discussed earlier in this report. So far, this has only been investigated in two dimensions because of gravity, for example, due to sedimentation of the active swimmers. Microgravity allows experiments in the appropriate three dimensions. We now have some artificial systems that show templated self-replication and exponential growth (He et al., 2017). When extended to the colloidal scale, it will be interesting to compare two-dimensional (2D) versus 3D growth. It would also be interesting to use microrheology to study the rheological behavior of polymers in the presence of cleavage enzymes in mesoscopic samples unsettled and undisturbed for long periods in microgravity.

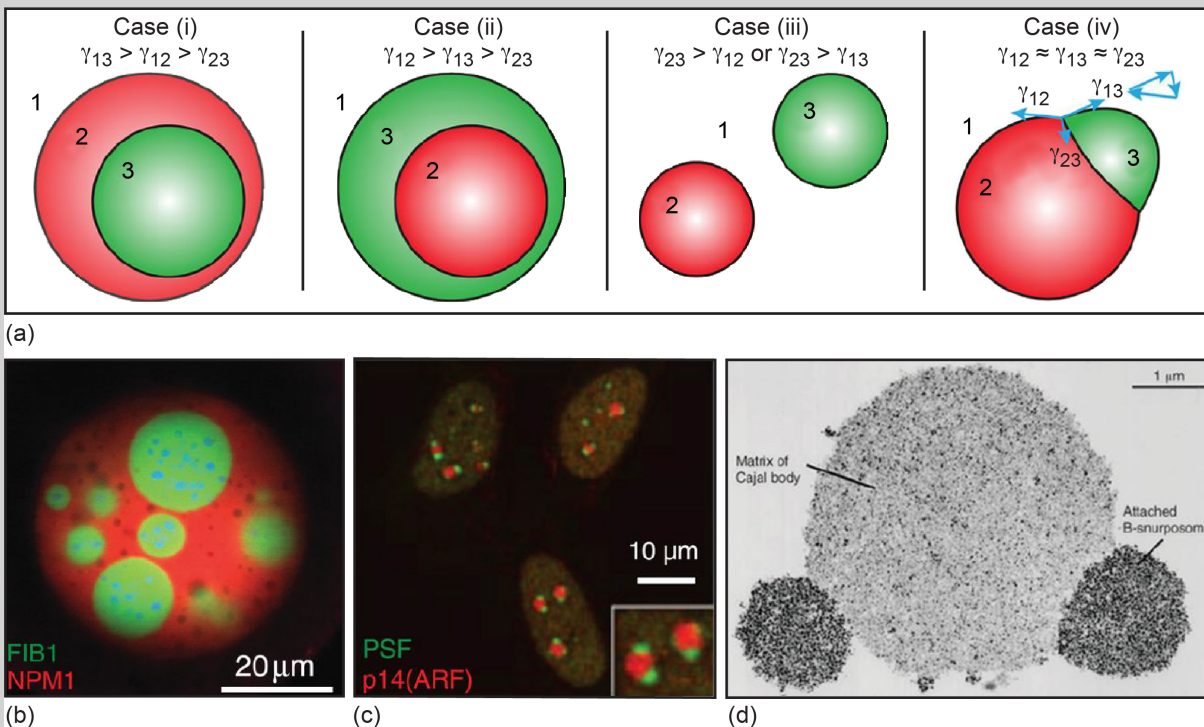


Figure 8.—Surface tension and multiphase droplet architecture. (a) Relative surface tensions among different possible interfaces ( $\gamma_{ij}$ ) dictate droplet architecture. Minimizing free energy of system requires minimizing energetically costly interfaces. For example, in Case (i) the costly 1–3 interface ( $\gamma_{13}$  large) is avoided by phase 2 enveloping phase 3, whereas Case (ii) achieves opposite. For Case (iii), interface between the two droplets is costly (high  $\gamma_{23}$ ), and thus, droplets do not contact one another. When relative energetic costs are comparable, all three phases can have shared interfaces, as shown in Case (iv). Surface tensions from three interfaces are balanced, forming Neumann’s triangle. (b) Multiphase nucleolus after actin disruption in *Xenopus laevis* nucleoli. (c) Transcription inhibition leads to reorganization of nucleolar architecture, forming perinucleolar caps bound to nucleolar bodies in HeLa nucleoli. (d) Electron micrograph showing *X. laevis* Cajal body with attached B-snurposomes. Adapted from Yongdae and Brangwynne, 2017.

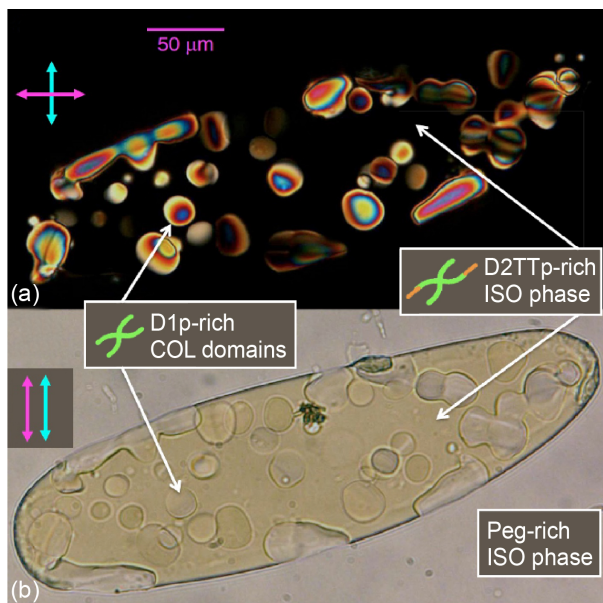


Figure 9.—Aqueous microdroplet reactor for liquid crystal (LC) autocatalysis of ligation of the deoxyribonucleic acid (DNA) 12-mers D1p (brightly birefringent under crossed polarizers). Duplex 12-mers D1p and D2TTp phase separate from polyethylene glycol (PEG) solution, forming yellow drop and then further separated into columnar (COL) LC domains of D1p (brightly birefringent under crossed polarizers) and dark isotropic (ISO) domains of non-LC-forming D2TTp. Having distinct LC and non-LC DNA phases enables comparison of ligation efficiency at same DNA density. Ligation is much more rapid in D1p LC fraction. Second phase transition takes place within DNA-rich droplet after it was already separated from PEG-rich ISO phase, appears from continuous and smooth interface with PEG-rich phase that enclose both DNA ISO and DNA LC domains. (a) DNA-rich droplet imaged under crossed polarizers. (b) DNA-rich droplet imaged under parallel polarizers. Adapted from Fraccia et al., 2015.

## 2.7 Active Patterning and Structure Formation—Self-Limiting Assembly, Actuation, and Integration

### 2.7.1 Bioinspired Soft Matter

Living organisms act as exquisite engineers capable of creating structures and functionalities that are unmatched by anything human made. The goal of bioinspired soft matter research is to uncover the fundamental design principles underlying these and other remarkable biological functions, and to use this knowledge to create a new generation of materials that are endowed with properties heretofore found only in living organisms. One set of challenges facing soft matter research is to develop synthetic equivalents of the neurotissue and muscle tissue and then to integrate the subsystems into a functional machine (Litschel et al., 2018). A second set of challenges concerns the self-assembly of complex and functional materials into large, but finite-size, superstructures. Living systems routinely achieve size-controlled

assembly, such as bacterial microcompartments like carboxysomes, and fibers with finite diameters like collagen. In contrast, most inorganic materials form unlimited structures like crystals, and synthetic approaches to size-controlled assembly lag far behind assembly in biology. Further, learning to engineer the self-assembly of synthetic self-limiting structures would enable scalable design of new functional and adaptable materials, such as paintable photonic coatings, injectable biomedical scaffolds, and capsids for drug and gene therapy. To accomplish this goal requires the development of a suite of synthetic building blocks that without requiring external control, undergo equilibrium self-assembly that terminates at a prescribed finite size. The colloidal building blocks must have arbitrary programmable shape on the nanometer scale, anisotropic interactions with ( $k_B T$ ) precision in control and measurement of thermal energy, tight angular specificity, and controlled flexibility. A further challenge is to form self-limited assemblies using nonequilibrium assembly. Such finite sized structures are ubiquitous in biology. This requires the development of particles that can change their on and/or off rates.

### 2.7.2 Spatiotemporal Patterns

Turing was the first to postulate that networks of diffusely coupled compartments containing chemicals undergoing nonlinear reactions are capable of spontaneous symmetry breaking that lead to self-driven oscillatory spatiotemporal patterns. The rich phenomenology associated with the Belousov-Zhabotinsky (BZ) oscillating chemical reaction serves as an example of the reaction-diffusion systems that Turing envisioned. A challenge facing soft materials science is to develop model experimental systems to study reaction-diffusion networks of phase oscillators in the Turing-Kuramoto framework and then to couple these reaction networks to mechanical systems. What is the general modeling framework for such chemomechanical systems? The new field of active matter describes the continuum mechanics of materials that consume chemical energy and generate stress and strain. Can this category of theory be extended to chemomechanics?

In order for networks of phase oscillators to emulate neural tissue, the following must be specified (i) the topology of the network, (ii) the boundary conditions, (iii) the initial conditions, (iv) the volume of each reactor, (v) the coupling strength, (vi) the identification of whether the coupling is inhibitory or excitatory, (vii) the coupling directionality, (viii) the heterogeneity in oscillator frequency, and (ix) the coupling strength (Litschel et al., 2018) as illustrated in Figure 10.

### 2.7.3 Prospects

Soft robotics is a highly interdisciplinary field that combines traditional practices in robotics and computer science with principles and practices from the life sciences, materials engineering, and chemistry. Unlike conventional machines and robotic hardware, soft robots are primarily composed of elastomers, fluids, and other forms of soft matter. Their composition enables them to mechanically deform and physically interact with objects without imposing large stress concentrations. While originally focused on pneumatic “artificial muscle” actuators for human motor assistance and tentaclelike continuum end effectors for robotic manipulation, the field has since expanded to include a wide variety of biologically inspired robots with a rich range of material architectures, functionalities, and applications.

What are the optimal viscoelastic properties of chemomechanical shape-changing gels? How do we wire up subunits consisting of pairs of coupled oscillators into complex devices? How can we preserve the functionality of a module when it is incorporated into a multicomponent machine so we can utilize plug-and-play assembly methods? Can materials learn, or even compute?



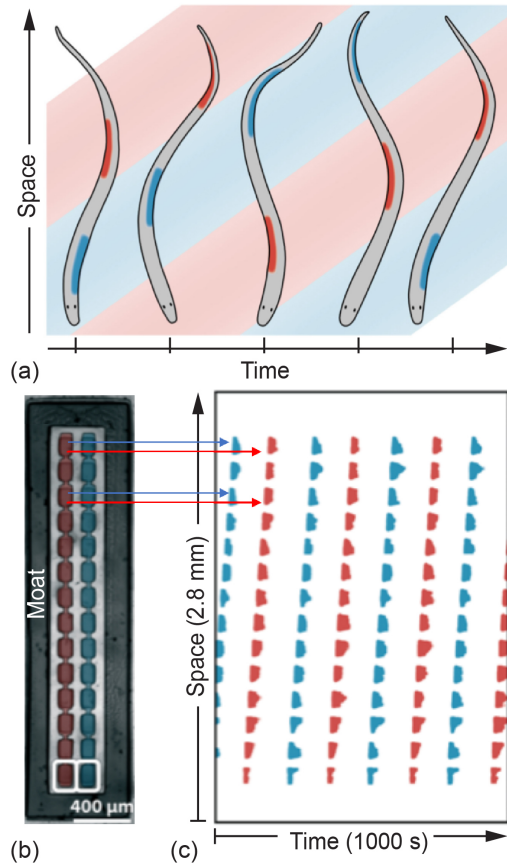


Figure 10.—Spatiotemporal activity.  
 (a) Space-time plot of swimming eel. Waves of contraction propagate down spine.  
 (b) Fabricated Belousov-Zhabotinsky (BZ) chemical oscillator network. (c) Measured space-time plot of BZ network. Blue records temporal activity from corresponding node of right side of (b), red the left side. Adapted from Litschel et al., 2018. Movie: <https://youtu.be/pi8wnXem-8o>

#### 2.7.4 Microgravity

We want to understand nonequilibrium structure formation and processing. As a start, it is worth focusing on a simple well defined case. As an example, apply uniform temperature gradients in soft materials such as gels and freeze in nonuniform stress across material. The fundamental aspect of response to a temperature gradient, thermophoresis, would benefit greatly from a microgravity environment that serves to isolate temperature gradients from other effects such as buoyancy-induced convection. As in other parts of this report, microgravity has the benefit of taking us from the 2D world dictated by sedimentation onto a surface to the 3D world that we want for useful fabrication. Microgravity also enables the study of fundamentally new forces such as fluctuation-induced Casimir forces that may prove useful for processing especially in space.

## 2.8 Fluids: Liquid Crystals

Having generated one of the trillion dollar technologies that enabled the portable computing revolution of the 20th-century, LCs face something of a “what have you done for me lately?” challenge. But this is an easy question to answer. LCs embody the broadest conceptual framework of soft matter, dealing directly with phenomenology at the interface of fluidity, and order, and ubiquitously manifest in soft matter systems and behavior including colloids, mechanics, rheology, polymers, glasses, hydrodynamics, granular materials, self-assembly, etc. Virtually every soft material has liquid crystalline features in its equilibrium and/or nonequilibrium states, connections that have driven LC research into the 21st century, providing exciting opportunities for new science and technologies (see Figure 11 for example).

Topology has been an important soft matter theme since LCs introduced the notions of topological defects, protection, and order into materials physics (in nematics in the 1920s), and the first technology based on topology (dynamic scattering in the 1960s). Currently, LCs are the leading experimental system for the detailed study, visualization, and discovery of novel topological behaviors, such as the recently developed paradigm for creating large families of colloidal atom inclusions of distinct symmetries in LCs (Senyuk et al., 2019). Experiment is paramount here: observation is revealing topological structures of complexity beyond imagination and current simulation capabilities, for example, 3D crystals of adaptive knots (Tai and Smalyukh, 2019). With respect to the latter, artificial intelligence emulation of large-scale simulations have demonstrated a reduction of  $\sim 10^9$  in computing time (Kasim et al., 2020), opening the way for far more effective simulation of collective equilibrium and active LC behavior at the mesoscale. Similar application of artificial intelligence emulation to atomistic or coarse-grained simulations will facilitate LC synthesis and molecular design.

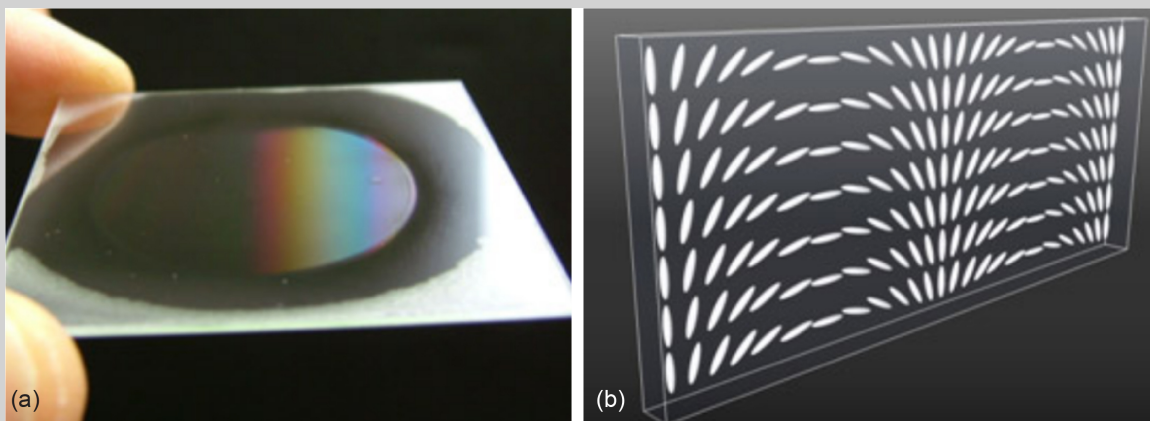


Figure 11.—(a) Prism providing  $15^\circ$  deflection of visible light beam made from few  $\mu\text{m}$ -thick liquid crystal (LC) film patterned in orientation into periodic cycloid director pattern shown in (b) (Pancharatnam/Berry) geometrical phase wave plate. Stacking plates increases deflection and circular periodicity makes lenses. Image from Beam Engineering, Inc. (<https://www.beamco.com/Cycloidal-Diffractive-Waveplate-CDW-Technology>).

### **2.8.1 Prospects**

New LC phases (dozens in some years) provide a fertile ground for broadening and understanding the interplay of fluidity and order, especially the recently discovered macroscopically ferromagnetic colloidal nematic phases of magnetic nanoplates, an example of using anisotropic colloidal particles as mesogens to form new phases (Mertelj et al., 2013; Shuai et al., 2016). Features of particle shape, flexibility, and chirality all matter, and the field is now turning to making particle characteristics dynamic, intelligent, and active, tools that will enable new modes of self-organization as well as “thinking” materials. Also exciting are macroscopically ferroelectric thermotropic small-molecule nematics with almost perfect polar ordering (X. Chen et al., 2020).

LCs are at the heart of a geometrical phase revolution in optics, wherein micron-thick LC films ordered into periodic cycloid patterns generate high-performance, high-numerical-aperture optical elements. These and smart windows (e.g., those that electrically switch into mirrors) exemplify emergent LC technologies that will have 21st-century impact.

### **2.8.2 Microgravity**

The pursuit of active soft matter phenomena based on hierarchical structuring is a dynamic field, bringing renewed excitement to soft matter. Soft matter and biophysical science now meld in an impressive research landscape that excites and amazes in its achievement and potential, for example, employing intricate and dynamic soft-material structures and networks to mediate various materials functionalities in real time. Locomotion as a topic, for example, ranges from the study of slime molds, which use distributed sensing, actuation, and motility, in a soft matter context, to bacterial motility driven by active fluctuations and controlled by LC textures, and to LC elastomers, which exhibit large, reversible strain and can be patterned and combined with other materials to make 3D hierarchical functional composite structures for soft robotics and locomotion robophysics applications. Progress in 3D architectures from materials and devices with different densities and in understanding the assembly processes is greatly facilitated by a microgravity environment.

## **3.0 Concluding Remarks**

This report lays out only some of the potential directions for soft matter dynamics over the next two decades. It also lays out the role that gravity plays in the organization of the basic building blocks of matter. Not only will research on soft matter have tremendous application towards understanding its behavior in our terrestrial environment, but also potentially in other NASA programs such as planetary science, exploration, robotics, etc.

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## Appendix A.—Originally Planned Session for 2020 American Physical Society March Meeting in Denver

The following information is the originally planned session for the Grand Challenges in Soft Matter and Opportunities for Microgravity Research presented by the Division of Soft Matter (DSOFT) of the American Physical Society on March 3, 2020, which was canceled and became the virtual Zoom meeting held on Thursday, March 26, 2020, at 11:30 a.m. to 1:30 p.m. EST.

Bulletin of the American Physical Society

APS March Meeting 2020  
Volume 65, Number 1  
Monday–Friday, March 2–6, 2020; Denver, Colorado  
Session Index

Session J31: Workshop: Grand Challenges in Soft Matter and Opportunities for Microgravity Research

Focus

Sponsoring Units: DSOFT  
Chair: Paul Chaikin, New York University  
Room: 503

J31.00001: Hot topics and lukewarm opportunities for soft matter science up in the sky  
Invited Speaker: Roberto Piazza

J31.00002: Non-equilibrium behaviour of colloidal systems  
Invited Speaker: Daan Frenkel

J31.00003: Squeezing order out of disorder  
Invited Speaker: Stefano Martiniani

J31.00004: ESA microgravity program for Soft Matter research  
Marco Braibanti

J31.00005: Electric field driven aggregation of negatively and positively polarized particles in dilute suspensions  
Boris Khusid, Qian Lei, Ezinwa Elele

J31.00006: Structure and dynamics of a two-dimensional colloid of liquid droplets  
Christoph Klopp

J31.00007: Shapes and forms for 2D liquid crystal materials  
Zhengdong Cheng, Dali Huang, Ugochukwu Okeibunor

J31.00008: Model hard ellipsoids: the practical matter of producing them  
Andrew Hollingsworth, Paul M Chaikin, Lou Kondic, Alton Reich, Boris Khusid

J31.00009: Coarsening of two-dimensional island emulsions on smectic liquid crystal bubbles in microgravity  
Cheol Park, Eric Minor, Joseph E. MacLennan, Matthew Glaser, Noel Anthony Clark, Christoph Klopp, Torsten Trittel, Ralf Stannarius

J31.00010: Temperature-gradient-induced thermomigration in smectic liquid crystal bubbles and freely suspended films in microgravity  
Noel Anthony Clark, Cheol Park, Eric Minor, Joseph E MacLennan, Matthew Glaser, Torsten Trittel, Alexey Eremin, Kirsten Harth, Ralf Stannarius

## Appendix B.—Workshop Announcement

Workshop: Grand Challenges in Soft Matter and Opportunities for Microgravity Research (DSOFT)

Organizers: Paul Chaikin ([chaikin@nyu.edu](mailto:chaikin@nyu.edu)), Noel Clark ([noel.clark@Colorado.EDU](mailto:noel.clark@Colorado.EDU)), and Sidney Nagel ([srnagel@uchicago.edu](mailto:srnagel@uchicago.edu))

Dear Colleagues,

We would like to rekindle the Workshop: “Grand Challenges in Soft Matter and Opportunities for Microgravity Research” (DSOFT), an opportunity lost with the cancellation of the APS March Meeting in Denver as a precautionary measure against the spread of the COVID 19 virus. We invite you to participate in a remote, Zoom, reincarnation of the workshop to be held next week on Thursday, March 26, 2020 at 11:30 a.m. to 1:30 p.m. New York Time (starting 8:30 a.m. on the west coast, 10:30 a.m. Chicago, 3:30 p.m. UK, 4:30 p.m. EU).

The goals remain to identify the grand challenges in the rapidly expanding field of soft matter and to produce a road map for microgravity research that will guide NASA. The roadmap, along with new discoveries, will influence the future of the field and address the resources that will be needed. The workshop and roadmap will inform NASA funding for future research areas in soft matter and complex fluids and the development of instruments and facilities for these studies. Potential topics of interest for the workshop include:

- Active matter
- Aerosols and particulates
- Biological materials
- Colloids
- Emulsions
- Fluids
- Foams
- Gels
- Glasses and disordered matter
- Granular materials
- Liquid crystals
- Rheologies
- Surface phenomena

A previous NASA sponsored workshop in 2003 produced a report that laid the ground work for new instruments on the International Space Station, ISS, and for two decades of microgravity research (<https://ntrs.nasa.gov/archive/nasa/casi.ntrs.nasa.gov/20030112858.pdf>). The current workshop will endeavor to produce a similar roadmap, and will foster a lively exchange of new ideas as participants will want to see that the most exciting and current science is included.

You are invited to participate in the workshop and also to provide premeeting input to the workshop by describing what areas you would like to see discussed in the report including any specific microgravity-related experiments, concepts, observations, modeling, and theory. If you plan to attend, or provide input, please send an email to Paul Chaikin ([chaikin@nyu.edu](mailto:chaikin@nyu.edu)) and cc Noel Clark ([noel.clark@Colorado.EDU](mailto:noel.clark@Colorado.EDU)) and Sidney Nagel ([srnagel@uchicago.edu](mailto:srnagel@uchicago.edu)).



## Appendix C.—Contributing Workshop Participants

All the contributing workshop participants are provided in Table C.1.

TABLE C.1.—CONTRIBUTING WORKSHOP PARTICIPANTS

Participant	Affiliation	Participant	Affiliation
Irmgard Bischofberger	Massachusetts Institute of Technology (MIT)	Peter Lu	Harvard University
Cliff Brangwynne	Princeton University	Andrea Liu	UPENN
Michael Brenner	Harvard University	Tom Lubensky	UPENN
Jasna Brujic	New York University (NYU)	Lisa Manning	Syracuse University
Zhengdong Cheng	Texas A&M University	Vinni Manoharan	Harvard University
Xiang Cheng	University of Minnesota	Cristina Marchetti	University of California, Santa Barbara (UCSB)
Itai Cohen	Cornell University	John Marko	Northwestern University
Eric Corwin	University of Oregon	N. Menon	UMASS
John Crocker	University of Pennsylvania (UPENN)	William Meyer	University Space Research Association at NASA Glenn Research Center
Karen Daniels	North Carolina State University	Fyl Pincus	UCSB
Sujit Datta	Princeton University	Dave Pine	NYU
Benny Davidovitch	University of Massachusetts (UMASS)	Cynthia Reichhardt	Los Alamos National Laboratory
Tony Dinsmore	UMASS	Leif Ristroph	NYU
Michelle Driscoll	Northwestern University	Mark Robbins	Johns Hopkins University
Eric Dufresne	Eidgenössische Technische Hochschule Zürich	Jennifer Ross	Syracuse University
Doug Durian	UPENN	Tom Russell	UMASS
Seth Fraden	Brandeis University	Cyrus Safinya	UCSB
Margaret Gardel	University of Chicago	Omar Saleh	UCSB
Sharon Glotzer	University of Michigan	Chris Santangelo	UMASS
David Grier	NYU	James Sethna	Cornell University
Shura Grosberg	NYU	Kate Stebe	UPENN
Andrew Hollingsworth	NYU	Howard Stone	Princeton University
Peko Hosoi	MIT	Vincenzo Vitelli	University of Chicago
William Irvine	University of Chicago	Dave Weitz	Harvard University
Randy Kamien	UPENN	Tom Witten	University of Chicago
Boris Khusid	New Jersey Institute of Technology	Zorana Zeravcic	École Supérieure de Physique et de Chimie Industrielles, Paris Sciences et Lettres Research University
Jane Kondev	Brandeis University	Jun Zhang	NYU
Sungyon Lee	Texas A&M University	Alexandra Zidovska	NYU



## Appendix D.—Breakout Session Discussion Leaders

The breakout sessions and their discussion leaders are provided in Table D.1.

TABLE D.1.—BREAKOUT SESSION LEADERS

Session	Leader
Self-organization only possible far from equilibrium—machines making machines	William Irvine
Instrumentation	Dave Weitz
Suspensions, foams, emulsions, colloids, and granular materials	Karen Daniels
Packings, simulation, and big data	Lisa Manning
Mechanical metamaterials and topological soft matter: allostery and auxetics	Vincenzo Vitelli
Soft matter, bioscience and biotechnology	Fyl Pincus and John Marko
Active patterning and structure formation—self-limiting assembly, actuation, and integration	Seth Fraden
Fluids: liquid crystals	Noel Clark







