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The Promise of Soft-Matter-Enabled Quantum Materials

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The field of quantum materials has experienced rapid growth over the past decade, driven by exciting new discoveries with immense transformative potential. Traditional synthetic methods to quantum materials have, however, limited the exploration of architectural control beyond the atomic scale. By contrast, soft matter self-assembly can be used to tailor material structure over a large range of length scales, with a vast array of possible form factors, promising emerging quantum material properties at the mesoscale. This review explores opportunities for soft matter science to impact the synthesis of quantum materials with advanced properties. Existing work at the interface of these two fields is highlighted, and perspectives are provided on possible future directions by discussing the potential benefits and challenges which can arise from their bridging.

1. Introduction

Driven in large part by the current excitement about quantum information science in general, and quantum computing in particular, the area of quantum materials has seen substantial growth in recent years.[1–3] Similar to the early days of nanotechnology, currently everything "quantum" captures the imagination even of the public, as evidenced by product labels ranging from "quantum detergents" all the way to "quantum cars." The term "quantum materials" was originally used more specifically to refer to strongly correlated electron systems, but has since been broadened to encompass a huge landscape of materials with the common thread of having properties which emerge from nontrivial, quantum level effects: spin–orbit interaction, topological order, and quantum coherence among others (**Figure 1**).[1,4–6] This resultingly diverse field has seen an explosion in productivity over the past decade, fueled by such advances as the experimental realization of long sought-after particles/quasiparticles,[7–11] the achievement of room-temperature superconductivity at high pressures,[12] the development of material pathways to realizing qubits for quantum computing,[13] and the discovery of emergent behavior in novel material systems.[14–16] Most importantly, the emergent properties of quantum materials show potential for transformative applications in a number of technological sectors that will impact our daily life. Examples include quantum computers with processing abilities many times more powerful than those available today, cybersecurity and encryption that would be unbreachable by today's standards, a national grid with almost no loss, or new personal and commercial electronics with dynamic on-demand properties.[2]

Coined by Feynman in a speech advocating the further development of science at small scales, the phrase "there is plenty of room at the bottom" describes well the vast majority of approaches to quantum materials. Progress in quantum material discovery and development has so far primarily relied on atomic scale synthetic methods, often with stringent conditions.[17] Increasingly sophisticated techniques in crystal growth and nanofabrication have given rise to the "age of silicon," and directly enabled the systematic study of quantum materials.[17] Conventional synthetic methods for quantum materials like molecular beam epitaxy (MBE), pulsed laser deposition (PLD), or single-crystal growth techniques often require ultrahigh vacuum or expensive equipment, however, which can impede widespread adoption and limit throughput. Furthermore, these traditional methods typically lack the ability to control structure beyond the crystal lattice, and are often limited in their range of available form factors.

Much untapped scientific potential lies in the expansion of quantum materials beyond the confines of these traditional synthetic techniques. In this context, the convergence of hard and soft condensed matter science opens up a particularly large opportunity space. Soft matter provides a wealth of systems with additional degrees of freedom for controlling structure over a wide range of length scales (**Figure 2**).[18] Basic soft matter synthetic elements, or "synthons" to borrow a term from organic chemistry, encompass a literally infinite set of building blocks with sizes well above the atomic scale that dominates the crystalline state of traditional quantum materials (Figure 2,

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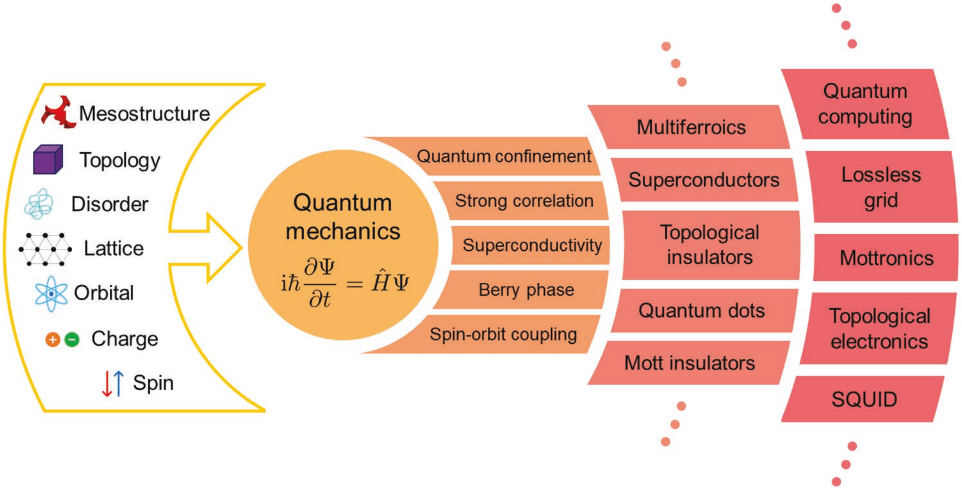

Figure 1. Quantum materials from fundamental system parameters (left) all the way to applications (right). System parameters are the different degrees of freedom that, upon treatment by quantum mechanics, give rise to quantum phenomena underpinning classes of quantum materials. With soft matter, we can identify additional degrees of freedom (e.g., disorder and mesostructure) expected to enable further altering and tuning of quantum material behavior.

bottom row). A salient feature of soft matter is the ability for these synthons to spontaneously self-organize into a plethora of mesoscopic periodic structures, a process generally referred to as self-assembly. The lattice parameters of these structures typically span a wide range from a few nanometers to several micrometers, well above those of atomic lattices (Figure 2, middle and top).[18] Most interestingly, and in stark contrast to conventional crystalline inorganic materials, in some of these self-assembled soft materials, lattice parameters of specific periodic structures can be tailored over a wide range on the mesoscale, e.g., by varying polymer molar mass via living polymerization techniques.[19–22] Synthetic routes to self-assembled materials are often referred to as "bottom-up" approaches, which stand in contrast to so-called "top-down" strategies including lithography, where materials are structured from the macroscopic scale downward.

The word "meso" originates from the Greek language and means "middle." The mesoscale, therefore, sits in-between

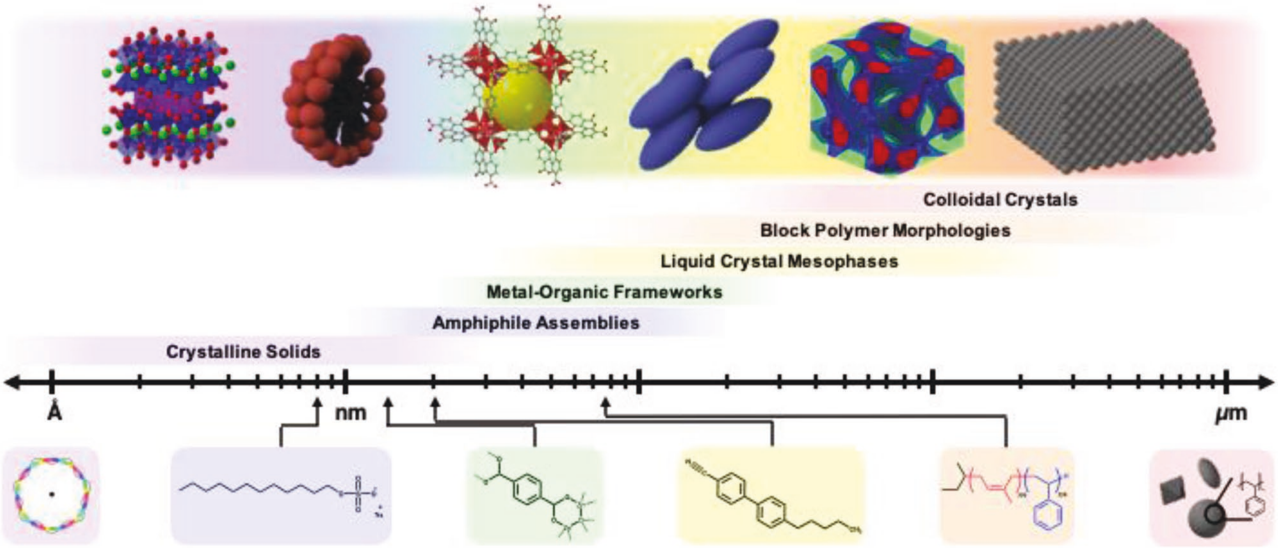

Figure 2. Comparison of the size of various synthetic elements, called synthons (bottom row), the length scales which can be reached by their respective periodic structures (middle row), and representative images of self-organized materials (top row). False coloring is used to connect synthons with their assembled structures, and represents the decrease in free energy of associated assemblies from left (small length scale, high energy) to right (large length scale, low energy). Bottom row from left to right: De Broglie representation of an atom, sodium dodecyl sulfate small molecule amphiphile, representative unit of MOF synthons benzenedicarboxylate and tetrahedrally coordinated Zn^{2+}, liquid crystal synthon (mesogen) 4-cyano-4′-pentylbiphenyl (5CB), poly(isoprene-*b*-styrene) block copolymer, and representation of colloidal polystyrene sphere, octahedron, or ellipsoid. Top row from left to right: yttrium barium copper oxide ($YBa_2Cu_3O_7$, YBCO) crystal structure, amphiphile-based spherical micelle, MOF-5 unit cell, representation of a nematic liquid crystal phase, single unit cell of a block-copolymer-derived core–shell double gyroid, and close packed colloidal crystal.

the atomic and macroscopic scales. The mesoscale is a fertile ground for the exploration of emergent quantum effects. For example, take the change in properties due to confinement. In the most basic sense, many quantum materials arise from interfacial phenomena, proximity, or reduced dimensionality.[4,7,23] Thus, the ability to tune interfaces or confinement on the mesoscale of nanometers to micrometers can have direct consequences on quantum material properties.[24] Superconducting nanoparticles experience an enhancement in critical parameters as compared to their bulk counterparts.[25,26] In HgTe/(Hg,Cd)Te quantum wells, there exists a threshold thickness of 6.3 nm across which the topologically trivial insulating well starts to harbor edge states, signaling topological phase transformations.[27] Attaining room temperature thermal stability at the 10 nm length scale has been identified as a key challenge for magnetoelectrically coupled multiferroics.[28] An attractive feature of soft-matter-derived network structures in particular is the ability to tune confinement in such materials while maintaining 3D continuous connectivity. This could offer opportunities to exploit effects, such as blueshifting of semiconductor bandgaps or enhancement in density of states, which arise from 1D-like confinement, while maintaining desirable, more 3D-like transport or other properties.[29] Furthermore, in some quantum materials, it is mesoscale characteristic lengths or structures which directly determine behavior. A prime example of this is in superconductors, where characteristic lengths which govern collective behavior of Cooper pairs and magnetic vortex behavior can range from a few nanometers to micrometers.[30] Common to other soft materials is the presence of photonic length scale structures for the study of light–electron or light–matter interactions.[31] Another effect which should be considered when shifting focus to larger periodic length scales is the resultant decrease in the frequency domain. This is exemplified very well in the exciting recent detection of topologically protected Weyl points in a macroscopic gyroid photonic crystal sample using microwave transmission experiments.[10] The construction, testing, and analysis of this material were made much more feasible by its increased periodic building block size. Similarly, other topological materials such as photonic topological insulators show much promise in addition to their quantum level, electronic analogs.[32,33]

Moreover, soft matter self-assembly can be used to structure direct other, e.g., inorganic materials lacking this ability for spontaneous self-organization. This enables efficient transfer of diverse and often complex sets of mesostructures, e.g., to more traditional quantum materials, thereby enabling emergent behavior. Such bottom-up self-assembly approaches have successfully been demonstrated across very different inorganic material classes, from amorphous glasses to crystalline transition metal oxides, and from semiconductors across metals all the way to superconductors.[34–39] The enormous amount of structural diversity obtained via self-assembly processes, and the ability to transfer this diversity into relevant "hard" materials, thus makes possible a plethora of novel synthetic routes toward quantum materials otherwise entirely inaccessible by traditional means.

Finally, as an additional benefit, soft matter processing regularly entails facile and cost-effective solution-based approaches, which typically can be scaled up in straightforward ways. Techniques like spin-coating or doctor blading, roll-to-roll (R2R) processing, and additive manufacturing or 3D printing are commonly used in soft matter design and manufacturing, but until today are largely absent in the study and use of quantum materials. Such approaches make soft-matter-enabled quantum materials particularly appealing for translation into the commercial sector. It is the combination of the enormous scientific as well as technological promise that motivates this rapidly expanding and highly interdisciplinary research area.

The self-assembly of soft matter systems has been employed in ever more sophisticated methods for the creation of periodically ordered materials on the mesoscale, but as yet this toolkit remains vastly underutilized in the synthesis and study of quantum materials.[24] While this will inevitably present new challenges for both the soft and hard condensed matter community, there is much potential for both fundamental scientific discovery and innovation by bridging the artificial separation, which has long divided these two fields. In this review, we hope to showcase this immense potential and demonstrate that, through soft-matter-enabled quantum materials, "there is plenty of room in the middle."

2. Progress in Soft-Matter-Enabled Quantum Materials

The twin forces of variety and volume make the prospect of efficiently categorizing the extensive work on quantum materials in conjunction with the even bigger field of soft matter into a comprehensive review a daunting task. As such, in this review, we shall follow the example outlined in the 2016 Department of Energy Basic Energy Sciences Workshop Report on Quantum Materials for Energy Relevant Technology of grouping materials based on the nature of their most apparent emergent behavior into three primary categories: superconductors, topological materials, and magnetic materials.[2] While it is inevitable that findings covered in this review will strain the bounds of any single category, we feel this to be a useful framework.

2.1. Superconductors

Superconductors have long loomed large in the public imagination. First discovered in mercury at low temperatures in 1911 by Heike Kamerlingh Onnes, one of the most salient features of superconductors is their ability to conduct electricity with zero resistance below a critical transition temperature, T_c.[40] The achievement of this resistance-free electrical transport at ambient conditions would obviously have enormous and far reaching technological impact, and therefore the realization of a "room temperature superconductor" has long been considered a holy grail of modern condensed matter physics. Central to the development of ever-increasing T_c is a firm understanding of the fundamental underpinnings of superconductivity. This understanding advanced first through phenomenological or macroscopic treatments, capped off by the holistic Ginzburg–Landau theory describing macroscopic superconductor behavior as a function of two characteristic lengths, the coherence length and the penetration depth.[40] The seminal work of Bardeen, Cooper, and Schrieffer (BCS)[41] later elucidated the microscopic mechanism of superconductivity, identifying supercurrent charge carriers to be electrons coupled by an attractive force into bosonic "Cooper pairs" which condense to form a macroscopic coherent state. In conventional superconductors, this attractive force is attributed to electron–phonon interactions.

Due to the limiting value of the Debye temperature, this phonon mechanism was predicted at the time to limit the T_c of strongly coupled superconductors of metals and metal alloys to a maximum of around 28 K.[42] This limit was far exceeded, however, with the surprising breakthrough discovery of superconductivity in mixed copper oxides;[43] excitement around this work led to the so-called "Woodstock of physics" at the 1987 meeting of the American Physical Society. Cuprates, including the popular yttrium barium copper oxide (YBCO) and bismuth strontium calcium copper oxide (BSCCO), opened the door to an entirely new category of technologically relevant high-temperature superconductors with T_c's above the boiling point of liquid nitrogen. Importantly, superconducting cuprates also joined organic Bechgaard salts[44] and heavy fermion superconductors[45,46] in the classification of unconventional superconductors, materials which could not be explained through electron–phonon coupling alone. Unconventional and high-temperature superconductors have since remained an extremely active field of study, with much work performed in the service of understanding these exotic materials[47] and achieving superconductivity in novel material classes.[15,48] The drive to achieve ever higher T_c's, however, stalled in 1993 at 133 K.[49] This record has only recently been exceeded, notably in conventional BCS type, hydrogen rich superconductors with high phonon frequencies and strong electron–phonon coupling under extreme pressures.[12,50,51] Exploration of this class of superconductors has recently achieved long sought-after room-temperature superconductivity; the record now stands at 287 K in carbonaceous sulfur hydride, albeit at a pressure of 267 GPa.[12]

A well-studied phenomenon in superconductivity is the effect of material confinement or structure on length scales commensurate with characteristic superconductor length scales.[25,40,52–54] Common considerations in granular superconductors include proximity effects,[55] Josephson coupling between individual grains,[52] quantum phase slips,[56,57] changes in phonon frequency,[58,59] size-dependent quantum oscillations,[60] vortex pinning forces at interfaces or mesoscopic structures,[61–63] and discretization of electronic energy levels.[64,65] The nano- or mesoscale shape, structure, or distribution in a granular superconductor can thus have profound effects on critical parameters and behavior. The elucidation of these effects is important for industrially relevant applications of superconductors, e.g., in composite materials, Josephson arrays, high T_c materials, and engineering magnetic behavior. Most explorations of low dimensional or confined superconductors have utilized traditional solid state synthesis methods.[25,66,67] A few efforts, however, have made use of more facile solution-based techniques, relying on organic moieties in order to control size and shape: small molecule ligands, surfactants, biomolecules, or polymers (**Figure 3**).

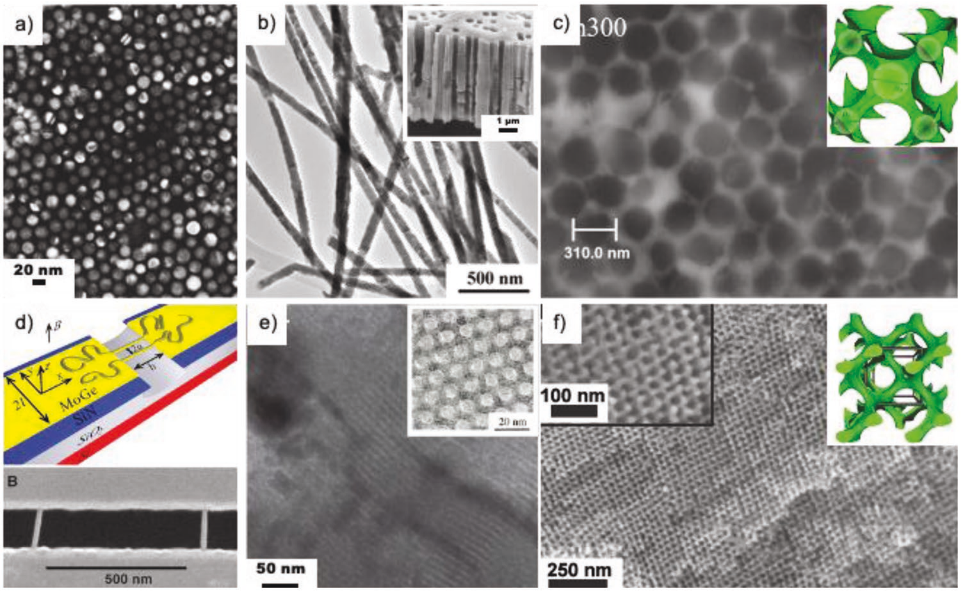

Figure 3. Superconductors of various ge ometries via soft matter self-assembly. a) Lead quantum dot superlattice. Adapted with permission.[68] Copyright 2000, American Chemical Society. b) Zinc nanowires grown via electrodeposition into a track etched membrane. Adapted with permission.[69] Copyright 2005, American Chemical Society. Inset: parent track etched membrane. Adapted with permission.[70] Copyright 2001, Elsevier Science Ltd. c) Tin inverse opal via melt injection of opal lattice; inset: depiction of the lattice geometry. Adapted with permission.[71] Copyright 2019, IOP Publishing Ltd. d) MoGe nanowires from DNA templates. Adapted with permission.[72] Copyright 2005, American Association for the Advancement of Science. e) Lead nanowires templated with SBA-15 silica. Adapted with permission.[73] Copyright 2013, American Chemical Society; inset: parent mesoporous SBA-15 silica. Adapted with permission.[74] Copyright 1998, American Association for the Advancement of Science. f) Block-copolymer-directed NbN with alternating gyroid structure; inset depiction of the alternating gyroid lattice. Adapted with permission.[39] Copyright 2016, the Authors, published by American Association for the Advancement of Science. From ref. [39]. Copyright The Authors, some rights reserved; exclusive licensee AAAS. Distributed under a CC BY-NC 4.0 license http://creativecommons.org/licenses/by-nc/4.0/. Reprinted with permission from AAAS.

This work spans quasi-0D nanoparticles, 1D nanowires or nanorods, 2D thin films, to bulk 3D mesoscopic architectures. In all these areas, mesoscale shape and structure have been revealed to intimately affect superconductor properties.

In the quasi-0D regime, much work has been performed to create nanoscale superconducting objects. In many studies, quantum dot synthetic techniques have been used to create isolated nano-objects or weakly Josephson coupled arrays of superconducting lead nanocrystals,[68,75–78] Pb/PbSe core shell particles,[79] gallium nanoclusters,[80] or palladium bismuth chalcogenide nanoparticles.[81] In much of this work, it has been found that critical field strength and critical temperature typically increase with decreasing material size until a certain material-dependent point, at which critical field, H_c, and T_c decrease until superconductivity is lost. This is consistent with seminal Anderson criteria,[82] and can often be understood at least partially in terms of increased phonon coupling strength via surface modes competing with an increasing average energy gap between successive electronic energy levels as material dimension is decreased, the so-called "kubo gap." With the claim as the first to demonstrate solution phase synthesis of monodisperse and stable lead nanoparticles, Weitz et al. were able to tune both particle size and proximity in the condensed colloidal solid (Figure 3a).[68] By decreasing ligand length from eighteen to six carbon chains, this work showed that superconductivity in the lead quantum dots was first suppressed, then localized to a few nanoparticles, then delocalized to the entire particle lattice.[68]

Proximity and shape can also be tuned in superconducting wires or wire networks in the quasi-1D regime through the use of strategies involving soft matter. Zhang et al. made use of sodium dodecyl sulfate for the soft templating of single crystal tin nanowires, and found that both the transition temperature and critical field strength were enhanced.[82] By tuning the ligand composition during growth, Moayed et al. synthesized superconducting "zigzag" lead sulfide (PbS) nanowires.[79] Bezryadin and co-workers used DNA molecules as templates for superconducting wires (Figure 3d).[72,84,85] Electrodeposition into track etched polycarbonate membranes has also very often been used to create nanowires[86] (Figure 3b) of ruthenium,[87] gold,[88] bismuth,[89–92] zinc,[69,93–96] tin,[97–105] lead,[97,99,105–109] or composites and alloys thereof.[110–112] Wire networks made in this way have often shown similar behavior as discussed previously in Josephson arrays of nanoparticles, and have been used as a platform to study the Little–Parks effect, phase slip centers, proximity effects, and other effects of nanostructuring on superconductors. A less common templating route to ultrathin nanowires is through the use of mesoporous SBA-15 silica, reported by Lortz and co-workers for both lead and niobium nitride.[73,113] He et al. found that the nanostructured lead confined to 6 nm porous silica (Figure 3e) switches to type-II superconductor behavior with evidence of long range order in 3D via Josephson coupling between parallel nanowires.[73] Wong et al., however, found that 5 nm NbN/silica nanocomposites behaved as a quasi-1D material, with no percolating bulk superconducting state observed at low temperatures.[113] This is attributed to a suppression of Josephson interactions between neighboring NbN nanowires due to the much smaller coherence length as compared to Pb.

Through bottom-up synthesis methods using self-assembled structures, a number of interesting phenomena have been observed in mesoscale ordered 2D superconductors. In the work of Vanacken et al., for instance, Nb thin films were grown atop polystyrene colloidal crystals in order to create artificial flux pinning lattices.[114] Ruiz-Valdepeñas et al. utilized a diblock copolymer to alter the domain texture in a thin film Nd–Co/Nb magnetic/superconducting multilayer structure.[115] Jang et al. created colloidal suspensions of superconducting nanosheets, which upon electrophoretic deposition formed an asymmetric membrane.[116]

The expansion of the field of mesoscale ordered superconductors into 3D has been somewhat limited by a scarcity of appropriate synthetic methods. Several routes to 3D mesostuctured superconductors have begun to be explored, however, which showcase the potential for many classes of soft matter to create novel superconducting materials. For example, in recent work, DNA origami was used to create nanowires[117] or a cubic superlattice of octahedral frames coated with niobium.[118] Through targeted design, DNA octahedrons were formed around gold nanoparticles, which then assembled into simple cubic superlattices. By first replicating the DNA struts of this structure in silica, then niobium, Shani et al. demonstrated the fabrication of a 3D array of Josephson junctions.[118] Through the use of the highly tailorable platform of computer aided DNA origami, this method shows much promise for achieving a wide array of possible structures and symmetries in mesostructured superconductors.

An alternative route, the most common so far to 3D mesoscale periodic architectures in superconductors, is via infiltration into self-assembled bulk colloidal crystals. This strategy has been employed to create inverse opal structured lead,[119–121] tin,[71,121,122] indium,[121,123] gallium,[121,124] bismuth,[125] lead/bismuth alloys,[121] and tungsten bronzes.[126] In the cases of classical type-I metals, the inverse opal or opal composite materials behave similarly to granular superconductors, with size-dependent changes in magnetization behavior and typically increased critical fields and temperatures. Aliev et al. observed magnetization behavior in lead inverse opals, which suggested the presence of novel fluxoid states and phase transitions in the nanostructured metal, including multivortex and giant vortex states.[119] Additionally, Little–Parks oscillations were seen with periodicities commensurate with the opal unit cell size. Bykov et al. observed similar lattice-size-dependent superconductivity in inverse opal structured tin (Figure 3c), and from the magnetization behavior constructed a vortex phase diagram.[71] Aliev later prepared opal structured Li_xWO_{3-y} and Na_xWO_{3-y} tungsten bronzes, and found evidence for localized nonpercolating superconductivity with a high transition temperature of 125–132 K.[126] Previous observations in these opal-type superconductors may suggest the presence of an artificially ordered flux lattice, but this was notably disproved by a study in 2021 by Lee et al. using small-angle neutron scattering which found only evidence for disordered flux arrays.[127]

An emerging method for the creation of 3D mesoscale architectures in superconducting materials is through the use of block copolymer (BCP) self-assembly to structure direct other materials in the bulk or in thin films. This has been demonstrated for both equilibrium and nonequilibrium structures. In the work of Robbins et al., triblock terpolymer polyisoprene-*b*-polystyrene-*b*-polyethylene oxide (PI-*b*-PS-*b*-PEO) was used to create mesoporous niobium nitride superconductors with cubic cocontinuous single gyroid network structure (Figure 3f, inset) via evaporation-induced self-assembly with sol–gel-derived niobium oxide sol nanoparticles and subsequent heat treatment under ammonia.[39] The mesoporous NbN was found to have incomplete flux exclusion at low field strength, and magnetization behavior suggested flux penetration commensurate with the gyroidal lattice constant. Thedford et al. also found that indium metal switched from type-I to type-II behavior when confined in a gyroidal ceramic template, which similarly suggested that the BCP defined architecture could be responsible for the creation of magnetic vortices and hard pinning centers.[128] Notably, the use of bulk block copolymer coassembly has been demonstrated as a versatile method for independently tuning morphology and characteristic lattice parameters across a large mesoscale range (from typically tens to hundreds of nanometers). In 2021, Beaucage et al. applied this tunability in the synthesis of niobium carbonitride (NbCN) superconductors.[129] In addition to creating materials of a quality parallel to traditionally synthesized NbCN (T_c of 16 K), this work found that mesoscale morphology induced by BCP self-assembly had an effect on the transition temperature of NbCN materials which appeared chemically identical via X-ray diffraction analysis.[129] Furthermore, BCPs give access to many morphologies otherwise difficult to obtain, including chiral cubic and networked structures, and enable a wealth of possible processing degrees of freedom. Yu et al. showcased this in the use of spin-coating and lithography to create area defined thin film NbCN superconductors.[130] In this work, it was further shown that BCP templated mesoporous NbCN materials possessed enhanced critical fields in comparison to dense analogs.[130] Moving to nonequilibrium-derived BCP-directed structures, in the work of Hesse et al., coassembly of a titania sol with the triblock terpolymer polyisoprene-*b*-polystyrene-*b*-poly(4-vinylpyridine) was used in tandem with nonsolvent-induced phase separation to create hierarchically porous, asymmetrically structured titania membranes. After careful thermal processing under ammonia, these titania membranes were converted into superconducting TiN. At room temperature, such materials displayed record level capacity retention and power density as electrochemical double-layer capacitors, and at low temperature could find application as superconducting membranes for gas separation.[131]

2.2. Topological Materials

The exploration of topological phases of matter started when Thouless et al. identified a completely novel scheme of classifying electronic phases[132] based on the topology of the electronic band structure.[133–135] They showed that the Hall conductivity in the quantum Hall effect for a 2D electron gas in a strong magnetic field is proportional to a topologically invariant number, now called the Chern number.[133,136,137] This topological invariant distinguishes the quantum Hall phase from a trivial insulating phase, and experimentally manifests in the form of Hall conductivity that is protected against disorder, and one-way transport of charges on the edge of the sample[138,139] – features that are not found in conventional insulators. Microscopically, the topologically nontrivial behavior of the quantum Hall state is a consequence of the quantum mechanical behavior of electrons in a magnetic field, in particular, the nonzero phase acquired by the electron wavefunction as it travels a closed loop in momentum space[136] (this is an example of a "geometric phase," which does not originate from wave propagation, but is instead a result of the curved geometry of momentum space). Because of this unconventional origin, topological phases of matter cannot be classified based on Landau's theory of broken symmetry[140] that is used to identify conventional phases of matter.

A few years after this discovery, it was then theorized that an external magnetic field is not necessary to realize a topological material.[141,142] The strong coupling between the spin and orbital angular momenta of electrons in certain materials is sufficient to create a topological state, called the quantum spin Hall state, that can be described using a spin-Chern number. These topological materials exhibiting quantum spin-Hall effects have conducting edges and an insulating bulk, earning them the name "topological insulators" (whereas the name "quantum Hall insulators" is more common for topologically nontrivial insulators based on the quantum Hall effects). Following these theoretical predictions,[27] the first spin–orbit-coupling-induced topological insulator was realized in thin films of HgTe sandwiched between CdTe.[143] In contrast to the quantum Hall phase, topological insulators are not limited to 2D structures.[144,145] 3D topological insulators that exhibit spin-polarized surface states have also been recently studied.[146] It is noteworthy that, in stark contrast to superconductivity, which was first accidentally discovered and then explained using theoretical models based on the quantum nature of interactions, topological insulators were first described theoretically before candidate materials were identified and tested.

One of the hallmarks of topological materials described above is the existence of unidirectional edge states that do not backscatter in the presence of defects. These edge states are guaranteed by the so-called "bulk-edge correspondence principle," which states that the net number of unidirectional edge states in a topological material is related to a topological invariant number that is evaluated from the eigenmodes of the bulk material.[146] In the quantum Hall phase, the external magnetic field breaks time reversal symmetry and generates unidirectional conducting channels on the edges of the sample that are unaffected by sample defects.[138] On the other hand, in the quantum spin Hall phase, two counterpropagating channels exist on the sample edges with opposite electron spin polarizations in the absence of an external magnetic field.[27] Since Kramers theorem for electrons prevents any interaction between the two opposite spins in a time-reversal symmetric system, these edge modes are not backscattered by nonmagnetic defects in the sample. Thus, it is time-reversal symmetry itself that guarantees the robustness of edge modes in topological electronic insulators based on the quantum spin-Hall effect.

Since the origin of the nontrivial topology in electronic band structures is due to the geometrical phase acquired by the electron wavefunction, which is a general phenomenon of wave physics, any wave-based platform can, in principle, be engineered to exhibit topological phases. This was first recognized by Haldane and Raghu,[148,149] who showed that an analog of the quantum Hall phase can be constructed for light in a hexagonal array of magnetized ferrites with a periodicity commensurate with the wavelength of light, called a photonic crystal. This unleashed a deluge of research work dedicated to realizing topological phases in various photonic,[150,151] acoustic,[152] and mechanical[153] wave-based systems. It must be mentioned, however, that these classical wave-based systems do not have the same properties as their electronic topological counterparts. This is because electrons are fermions, whereas photons and phonons are bosons which do not obey Kramers theorem and allow mixing of opposite spins.[32] As a result, while electromagnetic and photonic structures with broken time-reversal symmetry (e.g., magnetized plasmas and ferrites) can realize the equivalent of quantum Hall insulators, additional symmetries are needed to realize the analogs of quantum spin-Hall topological insulators.[32] Furthermore, these light-based topological insulators remain fundamentally more fragile to generic perturbations than their electronic counterparts.

Apart from spin–orbit coupling and external magnetizations, time varying periodic potentials can also result in a topological state. Dubbed a Floquet topological insulator, such phenomena were first shown by irradiating a quantum well with microwaves.[156] In photonics, this state was first emulated in a mesoscale structure consisting of honeycomb lattices of helical waveguides in fused silica.[33] This structure is time-invariant, but propagation along the helical waveguides acts as an effective “time” axis. The signatures of topological protection, such as backscattering-free edge state propagation, were indeed observed experimentally in this structure (**Figure 4**a–c).[33] The hexagonal (honeycomb) arrangement of chiral helices utilized in this material is especially relevant to soft matter[157] since it can be self-assembled, e.g., using BCPs.[158,159] Soft matter in the form of organic photoresists can similarly be used to construct nontrivial topological structures based on helical waveguides using direct laser writing.

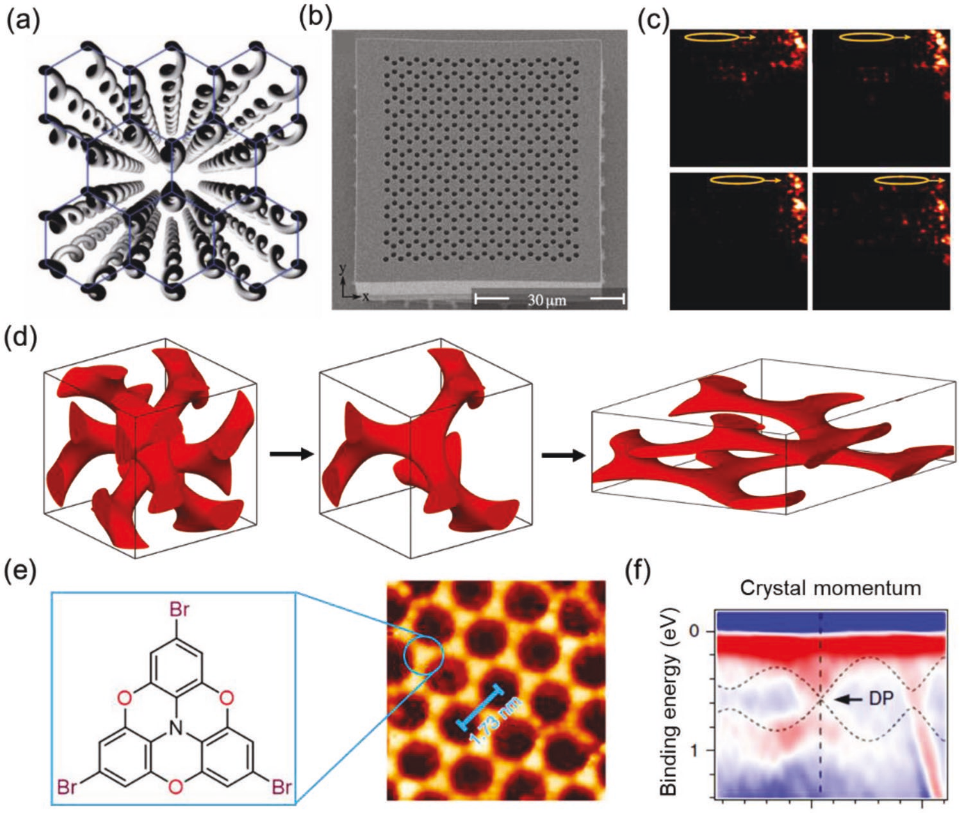

Figure 4. Topological materials from soft matter. a–c) Topological photonics. a) Schematic arrangement of an optical waveguide array composed of hexagonally organized helices, which implements a Floquet topological photonic insulator; b) image of the waveguide array structure made from an organic photoresist using direct laser writing; c) experimental demonstration of a topological edge mode propagating around the corner of the lattice with minimal backscattering (orange ovals indicate the irradiated regions). a,c) Adapted with permission.[33] Copyright 2013, Springer Nature. b) Adapted under the terms of the CC-BY Creative Commons Attribution 3.0 Unported license (https://creativecommons.org/licenses/by/3.0).[154] Copyright 2017, IOP Publishing and Deutsche Physikalische Gesellschaft. d) Symmetry breaking from double gyroid (left, space group *Ia*3*d*) via single gyroid (middle, space group $I4_132$) to orthorhombic networks (right, space group *F*222) to satisfy the symmetry requirements for the emergence of Weyl points (topological degeneracies in 3D momentum space) from self-assembled block copolymers. e,f) Dirac point (DP) in COFs. e) Molecular building block and the corresponding scanning tunneling microscopy image of honeycomb 2D structure; (f) second derivative angle-resolved photoemission spectroscopy (ARPES) data for observed DP with calculated bands (dashed lines). e,f) Adapted with permission.[155] Copyright 2020, Springer Nature.

Another topological phase of matter that was recently identified is the Weyl semimetal state.[160,161] Unlike topological insulators, Weyl semimetals are 3D materials and are not insulating in the bulk since the valence and conduction bands intersect at a point in 3D momentum space. The topological invariant characterizing Weyl semimetals can be determined from the gauge field corresponding to the phase acquired by the electron wavefunction, called the Berry curvature.[134] The points where the valence and conduction bands intersect, known as Weyl points, behave as sources or sinks of Berry curvature.[162] The topological charge of the Weyl point can be calculated by integrating the Berry curvature on a closed surface around the corresponding Weyl point in momentum space. Since the charge carried by the Weyl points is conserved, disorder or impurities in the material cannot create or destroy Weyl points, making the Weyl semimetal state exceptionally stable. Based on the symmetries of the Berry curvature, it can be shown that Weyl points can only be found in materials that have either broken inversion or time-reversal symmetry.[161]

In photonics, Weyl semimetals were first experimentally observed in a photonic crystal consisting of a double gyroid structure[10] that is also found in a wide range of assembled soft matter mesostructures. Recently, this structure has inspired a bottom-up approach of self-assembly of BCPs to prepare mesostructures that can host photonic and phononic Weyl points.[163] This is achieved by breaking the inversion symmetry of double gyroids by moving to single gyroids (also referred to as asymmetric double gyroids or alternating gyroids), which is another mesophase accessible from ABC-type triblock terpolymer self-assembly.[164] A further deformation of the single gyroid network lattice to achieve point group D2 symmetry is then required for the Weyl point to emerge. This symmetry reduction has recently been experimentally evidenced in hybrid thin films of heat-treated PI-*b*-PS-*b*-PEO structure directing resorcinol resols that are subsequently used in transient laser heating experiments to transfer the structure control into semiconductors like silicon (Figure 4d). The appearance of (002) peaks in grazing-incidence small-angle X-ray scattering of final nanostructured and mesoporous silicon suggested sample deformation-based symmetry breaking from space group $I4_132$ of a single gyroid upon solvent-evaporation-based self-assembly and subsequent thermal and laser treatments.[165] The facile and scalable processing via spin-coating, solvent vapor annealing (SVA), general furnace heating, and transient laser annealing paves the way toward photonic/phononic Weyl material fabrication from BCPs and other soft matter self-assembly. Furthermore, since all of these steps are consistent with semiconductor processing, direct integration into chip fabrication is also possible.

Compared to hard-matter-based topological electronic insulators, soft matter mesostructures that implement photonic/acoustic topological materials typically have a less-than-perfect structure and often dissipate the energy of the propagating photonic or acoustic wave in the form of scattering or absorption losses. This dissipation induces non-Hermitian perturbations to the Hamiltonian describing the system.[165] While this is often seen as a nuisance, as it prevents longer propagation distances, recent studies have shown that non-Hermiticity can introduce rich topological features not found in Hermitian systems such as exceptional points:[166,167] degeneracies where both eigenvalues and eigenvectors perfectly coalesce. In addition, exceptional points are associated with a local band dispersion that follows a square-root function around its branch point (in contrast to the linear dispersion of other degeneracies like Dirac points or Weyl points), which has been recently found to be potentially useful for enhanced sensing[168–170] and modal transformations.[171–173] Interestingly, the non-Hermitian nature of these systems does not necessarily alter the topological invariants of the system, which are preserved in the presence of moderate values of dissipation, but rather the specific impact of dissipation depends on the mechanism that makes the system topological, i.e., broken time-reversal symmetry or inversion symmetry.[174] Furthermore, at atomic length scales, self-assembled soft matter is typically amorphous; however, this also does not necessarily destroy the topological robustness of the system since topological edge states have been known to persist in the amorphous liquid regime.[175–177]

While photonic and phononic topological materials have been demonstrated using soft matter, topology has also increasingly begun to permeate soft-matter-derived materials based on electronic waves. 2D metal–organic frameworks (MOFs) have been predicted to possess nontrivial band topology.[179] MOFs are highly porous coordination polymers made from organic ligands stably coordinated to a metal center. MOFs can have 1D, 2D, or 3D structures, and their hybrid organic–inorganic nature can give rise to many exotic electronic and magnetic properties.[180] This transcends the notion that topological phases are restricted to purely inorganic materials.[181–183] The versatility of reticular chemistry in the synthesis of MOFs offers the freedom to perform chemical substitution at the molecular level, to modify the connectivity, and hence the lattice type, of the MOF network, and to tune quantum effects such as spin–orbit coupling by changing the metal ion. We expect the organic molecular linker, as with other soft matter, to be adaptive in that it can be engineered over a wide range of length scales and physically strained to the extent not allowed for atomic lattices. Indeed, linear degeneracies, called Dirac cones, have been revealed by angle-resolved photoelectron spectroscopy (ARPES) in 2D π-conjugated polymers (or covalent organic frameworks, COFs), whose lattice is an order of magnitude greater than that of graphene (Figure 4e,f).[155] In this scenario, certain perturbations that break inversion or time-reversal symmetry could then open a nontrivial topological gap in the spectrum, resulting in a transition to a topological phase.

In summary, we foresee that soft matter will play an important role in fabricating topological materials for photons, phonons, and electrons, for applications spanning different domains of physics and engineering. In addition, further explorations into the combined structure formation of both high-quality crystalline atomic lattices of topological electronic materials and the periodically ordered mesophases of topological photonic/phononic soft matter may open new opportunities at the intersection of different disciplines.

2.3. Magnetic Materials

Magnetism has been studied for centuries, with arguably the first published approach of scientific inquiry to magnetism being William Gerbert's De Magnete from the year 1600.[184] While inquiries into magnetism have ranged from its impact on nanoparticles all the way to planet earth, much recent work in this area has probed into quantum mechanics, elucidating exotic magnetic quantum materials.[184] In the broadest sense, magnetism as a quantum phenomenon is associated with the electronic spin degrees of freedom.[2] Magnetism has been used to produce a variety of interesting physical phenomena that hold great promise for and increase fundamental understanding of exotic systems,[2] as well as applications in areas including computing and energy.

Multiferroics are materials that simultaneously display more than one type of "ferroic ordering," i.e., ferromagnetism, ferroelectricity, and ferroelasticity.[28] Of particular interest for future applications in energy and "smart" technologies are magnetoelectric (ME) multiferroics, in which the materials are both (anti) ferroelectric and (anti)ferromagnetic, or super-paramagnetic, and the magnetic field affects the electronic properties and vice versa. There are multiple routes to multiferroics.[28,185] A common route is making heterogenous structures combining independently ferromagnetic and ferroelectric materials. While there are several approaches to achieve these heterostructures, one promising strategy involves making magnetic islands in a ferroelectric matrix; traditional "top-down" methods to this end include nucleating and growing magnetic islands randomly on a substrate with methods such as nucleation and growth, and subsequently backfilling or growing the ferroelectric matrix around the islands.[186] Lithography and epitaxy have been employed to overcome this random nucleation and try to impose a periodic order on the nanoislands.[186] However, these methods are still expensive and not friendly to high throughput processing. Soft matter has offered some promising alternatives in heterogenous multiferroics.

While lithography and epitaxy can provide periodic order and shape control, soft matter offers similar advantages, with the added benefit of solution processability. Ren et al. made a common heterogenous multiferroic of magnetic $CoFe_2O_4$ (CFO) and lead zirconate titanate using sol–gel methods and the diblock copolymer poly(styrene-*block*-ethylene oxide) as a structure directing agent.[187] Multiferroic nanocomposites made from block copolymers have also been fabricated by using one of the blocks as the ferroelectric component.[188] Terzić et al. made a lamellar multiferroic heterostructured nanocomposite using an ABA-type poly(2-vinylpyridine)-*block*-poly(vinylidene fluoride-trifluoroethylene)-*block*-poly(2-vinylpyridine) triblock copolymer (P2VP-*b*-P(VDF-TrFE)-*b*-P2VP), where the P(VDF-TrFE) block itself was ferroelectric and gallic-acid-modified CFO nanoparticles were segregated into the P2VP blocks[188] (**Figure 5**a–c). Exciton photovoltaic polymers have also been used to progress toward all polymeric multiferroics; Lohrman et al. developed a diblock copolymer of poly(3-hexylthiophene)-*block*-(3-hexylcarboxylate thiophene), which had an all conjugated backbone connecting two blocks with distinct electronic properties and crystallinities.[189] This allowed for high charge separation and excitonic ferromagnetism in a thin film made of the microphase segregated block copolymer, creating a material with a strong magnetoelectric effect and paving the way for organic excitonic multiferroics.[189] The promise of polymers as the future of heterostructured multiferroics has not gone unnoticed; a recent prospective published by Xu et al. highlighted polymer templating methods as a low-cost alternative for fabricating large-area, nanostructured thin films of complex metal oxides, particularly focusing on multiferroic nanocomposites.[190]

Another soft-matter-enabled heterostructured multiferroic was a core–shell multiferroic nanocomposite made from DNA-functionalized $NiFe_2O_4$ (NFO) and $BaTiO_3$ (BTO) that were tethered together by complimentary DNA oligomers[191] (Figure 5d–f). These particular nanocomposites were able to be directed into superstructures by a magnetic field, due to the reversibility of DNA hybridization. The core–shell nanocomposites showed magnetoelectric coupling, and the bulk composites produced upon sintering were shown to have stronger magnetoelectric coupling than a bulk composite made from randomly mixing NFO and BTO nanoparticles.[191]

Furthermore, magnetoelectric multiferroics can be realized with a single-phase material and soft matter has offered routes to such materials. BCPs have been used to structure direct single-phase multiferroic materials with enhanced properties. Quickel et al. employed the diblock copolymer KLE-22 and sol–gel chemistry to fabricate mesoporous bismuth ferrite thin films; the strain induced by the imposed mesostructure resulted in higher magnetoelectric coupling in these films as compared to epitaxial thin film counterparts with similar oxygen vacancy levels.[191] Moreover, one family of MOFs, the dimethylamino manganese formates (DMAMnFs) with perovskite-like structures, display multiferroic behavior that occurs through temperature-dependent order–disorder transitions[179,192] (Figure 5g–i). This induces MOF transitions from antiferroelectric to ferroelectric, and paramagnetic to antiferromagnetic states, respectively. Other types of MOFs can also exhibit multiferroicity.[193] One example is a 3D porous MOF structure with ethanol included in it as a guest molecule.[194] Due to order–disorder transitions affecting the orientation of the polar ethanol molecules, the $[Mn_3(HCOO)_6](C_2H_5OH)$ MOF underwent paraelectric to ferroelectric and paramagnetic to ferromagnetic transitions at 165 and 8.5 K, respectively.[194] The diverse and dynamic structures of MOFs, as well as the large variability of their chemical make up for each potential structure, render them strong candidates for soft matter that will have intrinsic emergent quantum properties in magnetics and other fields.

Spin ices are another class of magnetic quantum materials that display interesting properties. Spin ices are geometrically frustrated spins that with their magnetic moments follow the "ice rule," first described for the atomic placement of hydrogen atoms in water ice lattices:[196] similar to the "two-near" and "two-far" positions of hydrogen around an oxygen atom in crystalline water (from two hydrogens of the H_2O molecule and two hydrogens from two neighboring H_2O molecules, respectively), they consist of corner-linked tetrahedra of magnetic ions with nonzero magnetic moment each ("spin"). In the low energy state, two of these moments point outward ("two-out") and two point inward ("two-in") for each of the tetrahedra. Due to these magnetic frustrations, spin ices realize exotic properties such as magnetic monopoles and residual entropy, the latter just like in water ice, where the number of configurations conforming to the ice rule grows exponentially with system size, leading

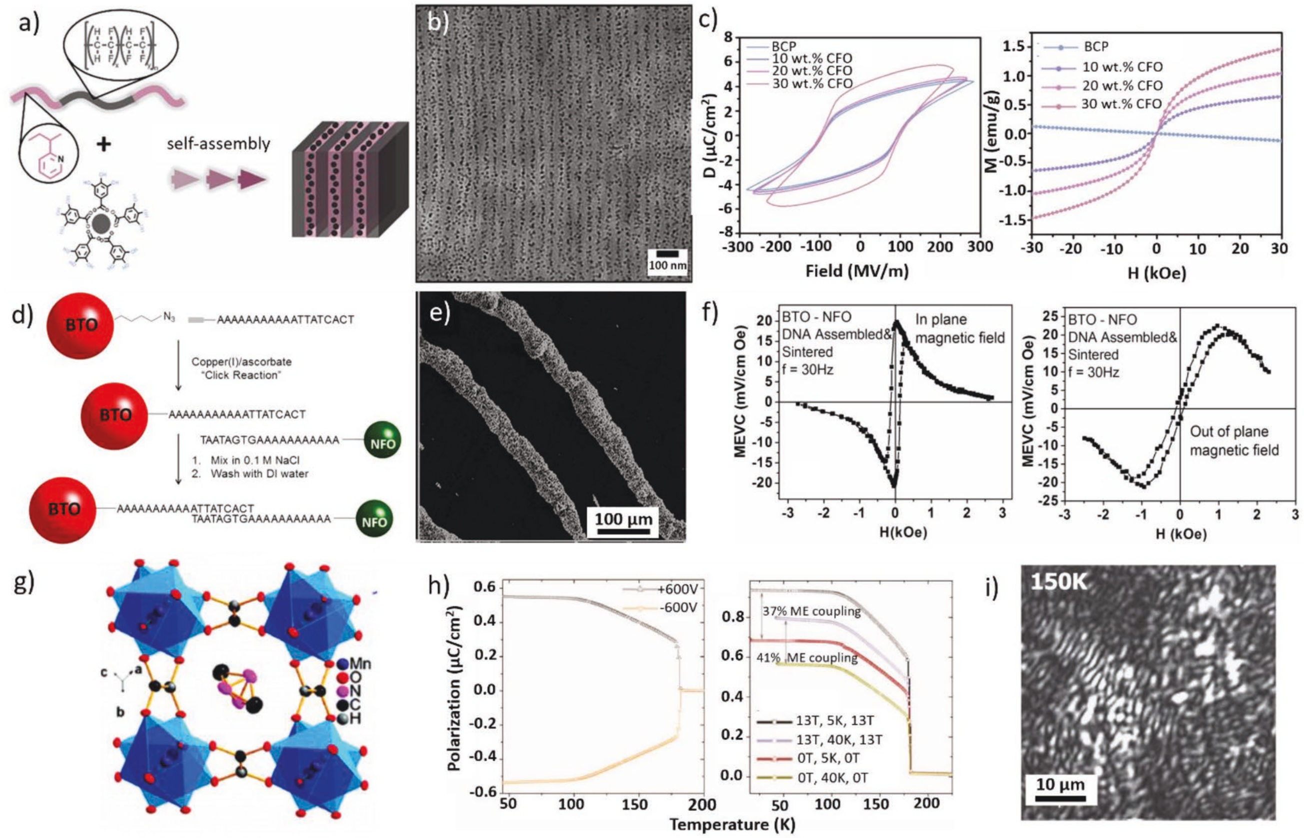

Figure 5. Examples of soft-matter-enabled multiferroics prepared from a variety of material classes: BCPs in the first row, DNA in the second row, and MOFs in the third row. a) Schematic of BCP with CFO NP coassembly to make lamellar multiferroics. b) Scanning electron microscopy (SEM) image of lamellar multiferroic made from coassembly of BCP and CFO NPs. c) Polarization and magnetic behavior of the BCP–CFO nanocomposite. a–c) Adapted with permission.[188] Copyright 2019, Royal Society of Chemistry. d) Schematic showing the DNA–DNA self-assembled BTO–NFO nanocomposites. e) SEM image depicting core–shell nanoparticles arranged into colloids by external magnetic field. f) Graphs of low-frequency magnetoelectric voltage coefficient versus in-plane and out-of-plane magnetic field, *H*, for sintered BTO–NFO DNA-assisted nanocomposites. d–f) Adapted under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://creativecommons.org/licenses/by/4.0).[191] Copyright 2016, Authors, published by AIP Publishing. g) Cartoon of the structure of DMAMnF MOF. h) Temperature-dependent polarization behavior of MOF, along with demonstration of the effect of external magnetic field on polarization behavior, demonstrating magnetoelectric coupling. Before measuring polarization, samples were cooled below T_c while applying a voltage of 600 V. The three numbers in the ME coupling legend provide, respectively, the magnetic field applied while samples were cooled, the target cooling temperature below T_c, and the magnetic field applied while measuring polarization. i) Optical image showing second-harmonic generation (SHG) in domains (bright areas) of the DMAMnF MOF, exhibiting areas that are non-centrosymmetric due to phase transition. g–i) Adapted under Creative Commons Attribution 4.0 International license (https://creativecommons.org/licenses/by/4.0).[180] Copyright 2016, Authors, published by Springer Nature.

to a substantial zero-temperature entropy. Typically, a spin ice is realized as nanomagnets at vertex points of periodic structures; this periodic nature leaves open a runway for soft materials to play a part in fabricating spin ices.[197] The spin ice was first discovered in rare earth titanate pyrochlores, which have a crystal structure with magnetic moments at the vertices of tetrahedra.[197] Attempting to replicate and control this phenomenon with synthesized materials led to the expansive field of artificial spin ices (ASIs). Through careful design and control of magnetic moments and emergent magnetic monopoles, ASIs have the potential to find applications in data storage, encryption, and both conventional and non-Boolean computation.[197] ASIs started as 2D square and kagome lattices with nanomagnets placed at vertices, but as the field developed, 3D ASIs were synthesized as well. One section of soft matter that has found much use in the field of ASI is colloidal crystals. Using magnetic particles and clever processing methods, colloidal scientists have generated a number of ASI and ASI analogs with colloidal crystals.[198–200] In 2013, Mistonov et al. fabricated an inverse opal-like structure of cobalt by backfilling a colloidal array of polystyrene microspheres; the resulting structure was able to follow the ice rule, making it a 3D ASI.[198] Further investigation of the micromagnetic structure in cobalt inverse opals via simulation and small-angle neutron scattering followed in 2019.[201] Ortiz-Ambriz and Tierno combined lithography with soft matter, using the colloidal self-assembly of paramagnetic particles in double-well traps to realize another colloidal spin ice.[199] In 2016, Shishkin et al. generated another inverse opal-like structure from a colloid, this time made from nickel;[202] their structure was considered a spin-ice analog since it was larger than considering lattice sites on an inorganic crystalline material.[200] Block copolymer templated architectures have also been investigated for controlled micromagnetic structure in 2011 by Hsueh et al. in nickel[203] and more recently in an in-depth study by Llandro et al. investigating Ni–Fe gyroidal networks.[204]

Many of the magnetic materials and physical effects described in the previous paragraphs find a particularly fertile ground within the broader field of spintronics, or spin electronics. This is a field of growing interest that uses the electron spin, in addition to its charge, in electronic devices for information technology. The associated extra degrees of freedom afforded by the electronic spin state, and the possibility of switching magnetic moments efficiently with spin-polarized currents, electric fields, and photonic fields, offer potential benefits including increased data storage density, reduced power consumption, and increased information processing speeds.[205,206] One early example of a commercially available spintronic device is the spin valve used in magnetic hard drives, which works due to the giant magnetoresistive effect, independently discovered by Fert and Grunberg in 1988, earning them the 2007 Nobel Prize in Physics.[206] Since these early investigations, the field of spintronics has been largely driven by the exploration of novel materials, including the already mentioned multiferroics, and there has been recent interest in investigating novel material systems for spintronics enabled by soft matter. For instance, biological soft matter has shown intrinsic properties than could allow them to play a part in molecular spintronics.[207] While PbS colloidal crystals have been known for their electronic properties, Moayed et al. demonstrated the potential value of colloids in spintronics.[208] The group was able to break the centrosymmetric O_h symmetry of PbS by confining the material to a 2D nanosheet and then subjecting the nanosheet to external asymmetric boundaries. The 2D nanosheet was sandwiched between the interface of SiO_2 and vacuum, and a gate electric field was applied; this resulted in the nanosheet of PbS belonging to the point group C_{4v} with no inversion symmetry. The resulting PbS colloid demonstrated spin–orbit coupling and the ability for solution-processible, industry-compatible colloidal crystals to be integrated into the growing field of spintronics.[208] Due to their highly tunable dimensionality, geometry, and chemistry, MOFs have also been investigated for applications in spintronics. For example, Chakravarty et al. used theoretical arguments and calculations to predict that bis(dithioline)-based MOFs have potential uses in spintronics and in realizing geometrically frustrated systems.[209] Beyond the first principal studies, Song et al. realized a semiconducting MOF thin film that was able to perform as a spin valve, using the MOF Cu_3(2,3,6,7,10,11-hexahydroxytriphenylene)$_2$.[210] The thin films were fabricated using layer-by-layer deposition, and had a magnetoresistance of up to 25% at 10 K.[210] Self-assembly of block copolymers has also begun to be used for the manipulation of spin waves, as shown by Manuguri et al. in the use of BCP micelles to direct a magnonic assembly (i.e., a material with magnetic properties periodically varied in space).[211]

3. The Convergence of Soft and Hard Condensed Matter

Over the past 30 years, the progression of soft matter science toward advanced functional materials provides illustrative examples of the opportunities and challenges for soft-matter-enabled quantum materials. Observing the convergence of soft and hard condensed matter physics over this timeline showcases several runways toward novel quantum materials in multiple soft materials classes (**Figure 6**). In soft matter, the characteristic length scale and morphology of mesoscale architectures is widely tunable via synthetic or processing means. Mesostructure is also adjustable via choice in soft matter agent, though notably a number of morphologies appear commonly throughout multiple materials classes due to some shared thermodynamic features.[212,213]

3.1. Molecular and Macromolecular Self-Assembly for Structure Direction and Templating

Self-assembling soft matter “synthons” can be used to induce nanostructure in other organic and inorganic materials that lack mesostructural ordering by themselves. This can be done through either structure direction, or soft or hard templating. Structure direction involves the cooperative assembly of a self-assembling “synthon” and other organic or inorganic precursor species. In particular, structure direction entails that the soft matter component and additive closely associate and assemble in tandem into intimately mixed hybrid phases. Precursor species can then be cross-linked or condensed in the resulting hybrid material, and the soft matter component removed via selective etching or heat treatment.[241] In 1992, Kresge et al. published a seminal paper using ionic amphiphilic surfactants to direct the structure of silica in solution into a periodically ordered mesoporous molecular sieve known as MCM-41 (Figure 6).[34] This paper can be considered as an important focal point for the use of soft matter self-assembly to structure direct inorganic materials.

Templating strategies also involve the use of a self-assembling soft matter component to impose a designer nanostructure on an inorganic or organic additive, but via routes which do not involve cooperative assembly as in structure direction. Templating strategies can be broadly classified into either “soft” or “hard” templating. Soft templating entails the use of “soft” materials, such as small molecule surfactants or polymeric species, which natively assemble into an ordered structure; organic or inorganic precursors then assemble around these soft templates and are transformed through various processing means.[241] The soft template structure is typically mechanically weak and prone to deformation, but can easily be removed via dissolution or heat treatment. An example of a soft templating approach can be found in the work of Thurn-Albrecht et al., where a self-assembled BCP thin film was used to template cobalt nanowires in a high density hexagonally packed array. After the neat diblock polymer film was assembled in a hexagonal array, one polymer phase was selectively etched out with UV light to create a porous polymer template in which cobalt was electrodeposited.[236]

By contrast, hard templating involves a rigid nanostructure made out of a material such as silica or carbon which already possesses an ordered mesostructure designed through earlier processes.[242] If such nanostructures are porous, the void space can be backfilled with various precursors which are then

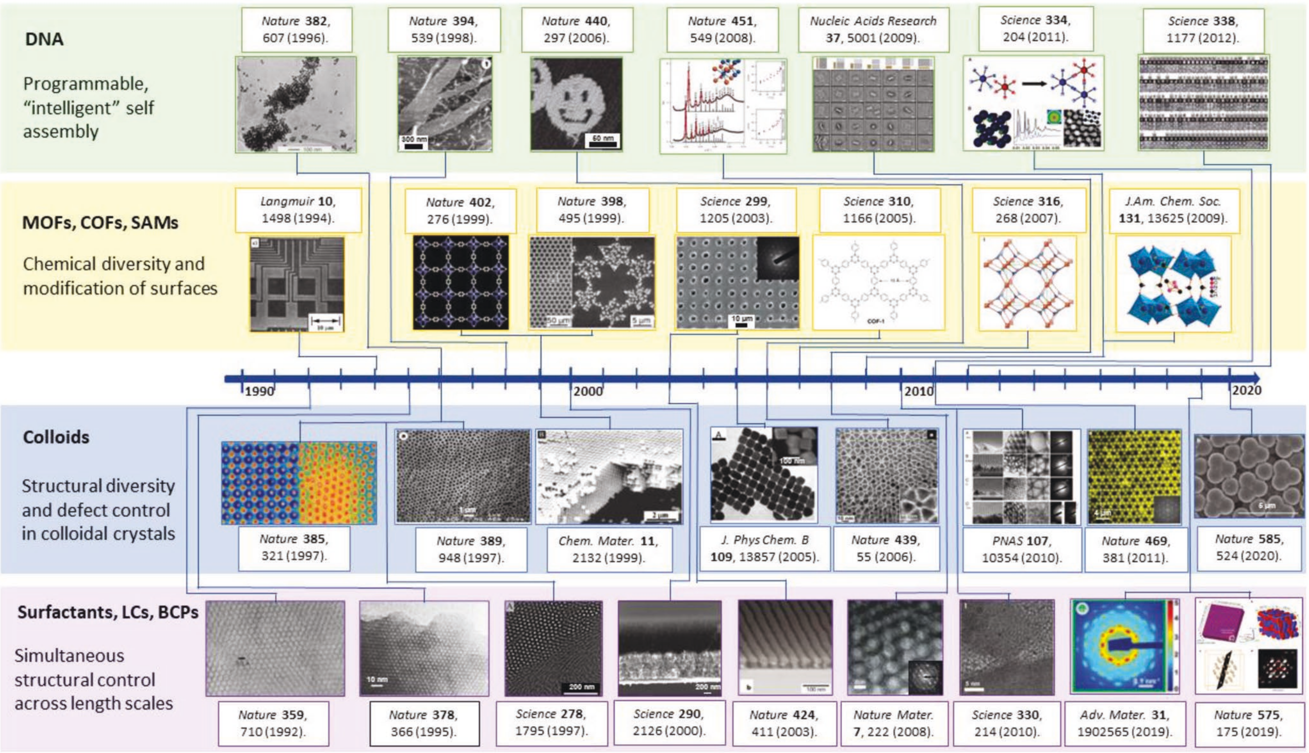

Figure 6. A timeline detailing seminal progress of the convergence of soft and hard matter across different material classes. The timeline shows a general trend of increasing sophistication of control over self-assembly, variety of materials being structure-directed, and quality of functional materials being structure-directed. DNA (top row) has shown progress from directing colloidal crystals of gold nanoparticles to intelligent, computer assisted design of DNA "origami." Top row (left to right): Adapted with permission.[214] Copyright 1996, Springer Nature. Adapted with permission.[215] Copyright 1998, Springer Nature. Adapted with permission.[216] Copyright 2006, Springer Nature. Adapted with permission.[217] Copyright 2008, Springer Nature. Adapted with permission.[218] Copyright 2009, Oxford University Press. Adapted with permisson.[219] Copyright 2011, American Association for the Advancement of Science. Adapted with permission.[220] Copyright 2012, American Association for the Advancement of Science. MOFs, COFs, and SAMs (second row) have introduced high levels of controllable crystallinity and intrinsic functionality into the field of soft matter.Second row (left to right): Adapted with permission.[221] Copyright 1994, American Chemical Society. Adapted with permission.[222] Copyright 1999, Springer Nature. Adapted with permission.[223] Copyright 1999, Springer Nature. Adapted with permission.[224] Copyright 2003, American Association for the Advancement of Science. Adapted with permission.[225] Copyright 2005, American Association for the Advancement of Science. Adapted with permission.[226] Copyright 2007, American Associaiton for the Advancement of Science. Adapted with permission.[193] Copyright 2009, American Chemical Society. Colloids (third row) have offered routes to the hard and soft templating of functional materials, with geometric variety ranging from packed spheres, to complex binary superlattices to kagome lattices.Third row (left to right): Adapted with permission.[227] Copyright 1997, Springer Nature. Adapted with permission.[228] Copyright 1997, Springer Nature. Adapted with permission.[229] Copyright 1999, American Chemical Society. Adapted with permission.[230] Copyright 2005, American Chemical Society. Adapted with permission.[231] Copyright 2006, Springer Nature. Adapted with permission.[232] Copyright 2010, The Authors, published by National Academy of Sciences, USA. Adapted with permission.[233] Copyright 2011, Springer Nature. Adapted with permission.[234] Copyright 2020, Springer Nature. Surfactants, liquid crystals (LCs), and block copolymers (BCPs, bottom row) have progressed from serving as a mere template for a few nanostructured silica and metal oxide mesostructures to sophisticated structure directing agents whose combined self-assembly with a variety of organic and inorganic precursors provides access to a plethora of mesostructured inorganic materials with varying symmetries and topologies as well as mesoporous materials with large surface areas. Bottom row (left to right): Adapted with permission.[34] Copyright 1992, Springer Nature. Adapted with permission.[235] Copyright 1995, Springer Nature. Adapted with permission.[35] Copyright 1997, published by American Association for the Advancement of Science. Adapted with permission.[236] Copyright 2000, American Association for the Advancement of Science. Adapted with permission.[237] Copyright 2003, Springer Nature. Adapted with permission.[36] Copyright 2008, The Authors, published by Springer Nature. Adapted with permission.[238] Copyright 2010, The Authors, published by American Association for the Advancement of Science. Adapted with permission.[239] Copyright 2019, The Authors, published by Wiley-VCH. Adapted with permission.[240] Copyright 2019, Springer Nature.

transformed into the final material. In such a way, the final inorganic or organic material adopts an inverse nanostructure to the template. The hard templates can endure harsher conditions such as high temperature and pressure, or corrosive and reactive environments during the templating and the following processing steps, which are not tolerated by soft templates under normal circumstances. However, selective removal of the template is often more difficult and requires harsh etchants. While one's intuition might not make the immediate connection between soft self-assembled materials and hard templates, using self-assembling materials to structure direct or soft template inorganic materials leads to a large variety of nanostructures and materials which can subsequently be used as hard templates. For example, Ryoo et al. used the small molecule surfactant soft templated silica molecular sieve MCM-48 as a hard template for the synthesis of ordered mesoporous carbon: the silica template was first created, filled with a carbon precursor solution of sucrose and sulfuric acid, heat treated in inert atmosphere, and then selectively etched away to yield a mesoporous carbon framework.[243]

At the smallest length scale, small molecules are known to form a variety of supramolecular architectures both in solution and the bulk, including spherical micelles, tubular micelles, lamellar sheets, and cocontinuous mesophases.[244–246] Liquid crystal (LC) mesophases appear in many systems of anisotropic organic molecules, termed mesogens.[247] While lacking the full 3D positional and orientational order typically defining a crystalline solid, liquid crystals possess limited degrees of orientational and positional order, which also distinguishes them from an isotropic liquid state. Lyotropic liquid crystals contain a mixture of an amphiphilic mesogen and solvent, and their phase behavior is governed by concentration. Thermotropic liquid crystals consist of neat mesogens, and their phase behavior is largely governed by temperature. The molecular structure of the mesogen, its size, rigidity, and asymmetry, plays a large role in determining the collective condensed phase assembly.[212,213,247] Understanding of the thermodynamic and structural constraints in this assembly allows for a fairly rigorous ability to describe mesomorphism via structural phase diagrams.[248,249] Characteristic lengths in small molecule self-assembly can range from nanometer diameter micelles to hundreds of nanometers in liquid crystal blue phases[250,251] or cholesteric chiral field periodicities.[252] Typically, the upper size limit can be substantially increased by the transition from small molecule to macromolecular building blocks.

Another class of soft materials that plays a significant role in opportunities for soft-matter-enabled quantum materials is the class of BCPs. BCPs consist of two or more chemically dissimilar, covalently linked polymer blocks. The wealth of existing techniques in polymer synthesis allow for an exquisite degree of control over many characteristics in these macromolecular synthons; tailoring of synthetic parameters leads to tunability in chemical composition, individual block length, and chain architecture.[253] Linear, cyclic, bottlebrush, dendrimer, and miktoarm star copolymers are all known to self-assemble into various periodically mesoscale ordered structures in which the morphology and mesoscopic unit cell are tunable over a wide range of length scales (Figure 2).[19] For example, in bulk linear AB diblock copolymers consisting of two immiscible blocks A and B,[254] mesoscopic morphologies ranging from close packed spherical micelles to the bicontinuous double gyroid[255] (Figure 2) can form depending on the relative volume fraction of the two blocks and the segregating strength between them. This phase behavior, sometimes referred to as a hallmark of soft condensed matter science, is understood to be due to the interplay of enthalpic contributions from interfacial energy and entropic effects from chain stretching, and can often be estimated from the molecular characteristics of a given polymer.[256] In more complex polymers such as linear ABC triblock terpolymers consisting of three chemically distinct blocks A, B, and C, the phase space can be quite rich;[257–259] additional blocks or other block architectures make this space even richer.[260] The ability to systematically design and produce a vast variety of BCP morphologies by synthetic control combined with the ability to tune the mesoscopic unit cell size via polymer chain length yields a powerful toolbox for the control of material structure on the mesoscale.

Like small molecule surfactants, BCPs offer pathways to mesostructured organic and inorganic materials through structure direction and templating strategies.[242] While low molar mass nonionic BCP surfactants have shown to structure direct silica in solution,[74] the thermodynamics of bulk BCP self-assembly has been used to structure direct inorganic materials in the condensed phase at larger length scales from higher molar mass copolymers, originally in the form of sol–gel-derived (alumino)silicate nanoparticles.[35] While phase behavior typically becomes more complex with inorganic precursors involved, when the correct design rules are being followed, the sequence of morphologies observed in the bulk adding more and more inorganic material can be very similar to that of the parent BCP.[242,261,262] This provides facile access to hybrid and mesoporous materials via evaporation-induced self-assembly (EISA) of bulk block copolymer mesophases, thereby coupling inorganic material structure formation with block copolymer bulk phase thermodynamics.[263,264] In addition to acting as structure directing agents, BCPs have been used as bulk soft templates to fabricate nanomaterials, as described earlier in the work of Thurn-Albrecht et al. among other examples.[236,241,265] BCP self-assembly derived ceramic molds have been used as hard templates to fabricate gyroidal networks of superconducting indium.[128] Furthermore, hard templates derived from BCPs have played a part in nonequilibrium transient processes; single crystal homo- and heteroepitaxial semiconductor and metal nanostructures, respectively, have been prepared using transient laser heating on the nanosecond time scale of materials that conformally backfill mesoporous BCP-directed templates.[238]

A strength of the chemical diversity of BCPs is that careful selection of blocks can allow for the combination of strategies to overcome obstacles. In 2008, Lee et al. used the combined assembly of soft and hard (CASH) chemistries method in order to successfully generate highly crystalline mesostructured metal oxides directed by bulk block copolymer self-assembly. Previously, mesostructured metal oxide crystals would outgrow the mesopores and collapse the structure during heat treatments to improve the degree of crystallinity. CASH chemistries involved the structure direction of metal oxides with a poly(isoprene)-containing BCP. The self-assembled hybrid material was first heat treated in argon, partially converting the sp^2 hybridized carbon containing poly(isoprene) into a hard carbon template that confined the metal oxide crystal growth and prevented mesostructure collapse; the resulting materials were then subsequently calcined in air, burning away the carbon and leaving behind the highly crystalline mesostructured metal oxides.[36] This example illustrates the versatility of soft-matter-enabled structure formation, here combining structure direction and hard templating in a so-called "one-pot" process, to make high-quality inorganic materials.

3.2. Colloidal Assemblies

Similar to the considerations describing macromolecules, colloidal self-assembly is rooted in intermolecular and microscopic forces, but scaled to particles comprised of many discrete atoms or macromolecules.[266] The combined effect of steric, dispersive, or Coulombic interactions of many constituent species creates attractive and repulsive interparticle forces. When these forces are properly balanced, colloidal particles self-assemble into close packed structures. In the simplest case, spherical nanoparticles will often organize into hexagonally closed packed structures much like artificial atoms.[267] More complex colloidal crystals can be obtained by coassembling particles of different sizes or surface charges.[231,268] While typically the interactions involved in such assembly are nonspecific, the introduction of complementary binding, e.g., via particle tethered DNA hybridization, can introduce a further level of rational design.[214,269,270] Beyond simple spherical particles, exotic structures can be created through the use of more complex colloidal building blocks and shapes: tetrahedra, ellipsoids, platelets, and so forth.[271–276] In all colloidal systems, the periodicity of close packed structures is dictated primarily by the size of the basic particle synthon(s). Synthetic advances have allowed for the creation of narrowly size-dispersed colloidal particles ranging from a few nanometers in size to multiple micrometers. Colloidal crystals thus cover a very broad range of length scales (Figure 2).

In the 1990s, colloids began to be used in templating methods to produce nanostructured inorganic materials from filling the interstitial space in emulsions or self-assembled polymer spheres (Figure 6).[276] Colloidal techniques were further refined by the innovation of colloidal epitaxy.[228] Colloids also saw a variety of creative approaches to new products in the 2000s. Colloidal crystals of a variety of exotic structures were produced by methods such as lost-wax approaches, using charged particles, or using binary particles of different shapes or sizes.[227,268,277] "Patchy" particles were also employed to self-assemble a colloidal kagome lattice from intermolecular forces in 2011 by Chen et al.[278] Recently, a similar strategy using deformed tetrahedral particles with tailored DNA patches accomplished the long sought after goal of a diamond structured colloidal crystal.[233] The progression in the field of colloid science once again highlights the potential of soft-matter-enabled quantum materials, e.g., via the templating of inorganic materials into deliberate, well-controlled, and increasingly sophisticated periodically ordered nano- and mesostructures.

3.3. Soft Interfaces and Coordination Polymers

Self-assembled monolayers (SAMs) were first generated from alkanethiols self-assembled into crystalline monolayer structures on gold.[279] Sometimes referred to as the "poor man's version of single crystal surfaces," they provide a large and versatile toolbox for the tuning and patterning of interfaces. For example, SAM surfaces can be chemically modified in myriad ways by controlling the chemistry of the alkanethiol headgroup.[221] In 1999, Aizenberg et al. employed this ability to control the nucleation of calcite crystals (Figure 6).[223] This control was subsequently extended by fabricating patterned SAMs used to generate large micropatterned single crystals of calcite,[224] thereby generating a large runway for the controlled formation of quantum materials at SAM-directed organic–inorganic interfaces.

As described in the previous section, MOFs and COFs also have a place in identifying routes from soft matter to quantum materials. MOFs are a type of soft matter that falls under the category of coordination polymers.[224] Generally, MOFs consist of one or more metal ions at the center, which coordinate to several polytopic organic linkers; this make up leads to a variety of geometries, depending on the metal centers and ligands used, as well as a variety of pore sizes of the final structure.[280] MOFs have been heavily studied for applications such as catalysis and gas storage since their inception due to their favorable properties such as high porosity, high crystallinity, and large chemical variety.[224,280] MOFs have such a variety of tailor-made properties because they exist as an exotic single-phase mixture of inorganic and organic matter.[224,280] In addition to the makeup of MOFs giving them a large spectrum of utensils from the toolbox of the period table, the variability in metal center and organic linker also leads to a vast array of possibilities for exotic structural topologies.[224] MOFs and COFs have well-controlled pores and crystallinity, as well as the ability to take on complex structural topologies in 2D and 3D.[224,281] The organic–inorganic hybrid characteristic of MOFs gives them exotic properties that make them more functional than many other classes of periodically organized soft matter. For example, as described in the previous section (Figure 5), MOFs in the dimethylammonium manganese formate family mimic perovskite-like structures and show multiferroic properties through phase transitions.[192]

3.4. Rational Design via Biomolecules

Another significant class of soft matter is associated with biological materials. Rational structural design employing DNA is a field of soft matter that began in 1982, thanks to the work of Seeman, which suggested that DNA could be used to make stable junction structures, and that those individual structures could be brought together through DNA base pairing to generate arbitrary nanoarchitectures.[283] DNA base pairs were subsequently used as tools for the assembly of nanoscale structures and materials, rather than just viewed as the basis for the genetic code, paving the way for the eventual rise of the field of DNA origami.[284] The use of these DNA base pairings to enable and control the assembly of matter at the nanoscale has since expanded from simple DNA–DNA interactions (Figure 6). In 1996, Mirkin et al. published seminal work on DNA oligomers being used to direct nanoparticles into colloidal crystals.[214] Biological soft materials based on DNA took another quantum step via the extremely impactful paper by Rothemund in 2006, directly combining the powerful structure directing capabilities of DNA origami with the use of computer algorithms for their structural design.[216] This paved the way to "hands-off" material formation all the way from computer-algorithm-based design to robotic synthesis and purification. Progressing further into the 2000s, the ability of DNA-assisted self-assembly to make increasingly crystalline and complex colloidal crystals has been investigated.[217,219] The progress in DNA self-assembly led to the advent of computer-aided design (CAD) in self-assembly.[218] This powerful tool has made it possible to use CAD software to explore self-assembly building bricks from DNA, opening up a world of possibilities in terms of structure control and its automated synthesis (as opposed to more conventional synthesis often guided by trial and error).

The past few decades have seen the field of soft matter develop such that a variety of organic and inorganic chemistries can be tuned across the nano- and mesoscopic scales in a multitude of geometries and periodic lattices. Progression in soft matter techniques has given rise to increasingly well-controlled and well-ordered materials, slowly beginning to move in the direction of the quality achieved in electronic materials. Crucially, high quality samples enable sophisticated measurements, e.g., of electronic and magnetic behaviors necessary for identification, characterization, and understanding of novel quantum phenomena. While there are many opportunities afforded by considering soft matter routes to quantum materials, there are therefore also challenges which will need to be addressed. The foremost challenge is to achieve, in soft matter systems, the same level of structural control available in traditional solid state or atomistic synthetic methods. Defects, grain boundaries, and disorder all can greatly impact measurements and obscure intrinsic properties. For example, grain boundaries have been shown to limit supercurrents in high-temperature superconductors.[285] The challenge posed is significant, but much work has already begun tackling the characterization and elimination of mesoscale defects. Coming back to the example of the fascinating cubic cocontinuous gyroidal network structures, in 2019 Feng et al. utilized “slice and view” scanning electron microscopy (SEM) to quantitatively characterize topological defects in the gyroidal morphology of block copolymers.[286] In the same year, Susca et al. produced the first macroscopic single crystals of gyroidal block copolymers with grains on the scale of millimeters.[239] Should the challenge of long-range periodic mesoscale order, as well as associated challenges in orthogonal device fabrication and measurement be met for a broad range of materials, soft matter is poised to open up a vast realm of new scientific and technological pursuits. Impactful contributions of soft matter are already being made, but there is plenty more room in the middle.

4. Comparing Traditional and Soft-Matter-Enabled Quantum Material Fabrication Techniques

A unifying concept in all soft matter systems is that assembly and behavior are governed by interactions of a similar magnitude as room temperature thermal energy. Individually weak hydrogen bonding, hydrophobic, screened electrostatic, or entropic forces collectively dictate mesoscopic structure and macroscopic properties. As a result, intermolecular or interparticle interactions are easily disrupted by low energy perturbations, but can just as easily recover. This leads to a substantial pathway complexity from synthons to the final self-assembled structure, with many possible metastable intermediate order states that may get trapped on the way to the lowest energy structure. At the same time, as energy minima are shallow, intrinsic structural variability can be substantial, and orientation distribution functions used to characterize molecular order in soft matter may be rather broad. Moreover, ordered states can be strongly influenced, e.g., by external mechanical, electrical, or magnetic fields, making the final structure exquisitely sensitive to material processing history, e.g., in injection molding or 3D printing. Most importantly, soft materials like polymers, colloids, and liquid crystals all are solution processible and therefore typically enable large-scale fabrication methods including, e.g., R2R processing and 3D printing from solution-based precursors. In turn, this flexibility in material processing leads to a wealth of possible form factors unavailable to traditional quantum material synthesis. This can readily be inferred from a comparison of some typical form factors derived from both traditional “hard” and “soft” material synthesis methods (**Figure 7**).

4.1. 2D Materials

Starting with graphene,[293] the family of 2D materials has grown rapidly to include a wide array of materials such as transition metal dichalcogenides, hexagonal boron nitride, and layered oxides.[287,294,295] These single layers can also be stacked together to form diverse and intimately controllable heterostructures with interlayer van der Waals bonding.[296] 2D materials and van der Waals solids have shown a plethora of exciting and exotic quantum materials properties such as 2D superconductivity, proximity-derived effects, and novel states in Moiré patterned multilayers.[297,298] Mechanical exfoliation provides a relatively facile and cost-effective means for obtaining many 2D materials, but lacks capability for high throughput. While much progress has been made on solution-based methods, 2D and van der Waals materials often rely on chemical vapor deposition under ultrahigh vacuum or versatile yet difficult layer-by-layer directed mechanical assembly.[299,300]

In soft matter, an equivalent to 2D materials in hard condensed matter physics lies in 2D polymers. 2D polymers are a scaled-up analog of atomic 2D materials such as graphene and transition metal dichalcogenides. While solid state chemical reaction from evaporated constituents is reported, 2D covalent organic frameworks or metal–organic frameworks have been successfully synthesized via low-cost, high-throughput solution reactions.[301] The soft matter constituent, small organic molecules in this case, is amenable to modification of chemical composition, linker size, and coordination geometry, all of which are severely restricted for atomic 2D materials. By perfecting the 2D structures, 2D polymers are a tunable form of soft matter awaiting interrogation of their quantum behavior and properties.

Also on the 2D level, SAMs organize on substrates into a highly crystalline form in a self-limiting way.[302] Such single molecular layers can tune the surface energy, useful for controlling crystal nucleation.[223] In a similar fashion, polymer brushes are another type of surface-tethering soft matter, though they usually functionalize the substrate with an amorphous layer. In addition to changing molecular identity as in SAMs, polymer brushes can be designed with different chain compositions to match the target surface energy, e.g., to prepare a neutral surface to avoid preferential wetting.[303] Surface-functionalizing soft matter thus offers opportunities for both the study of quantum phenomena in confined dimensions and the fabrication of novel quantum materials.

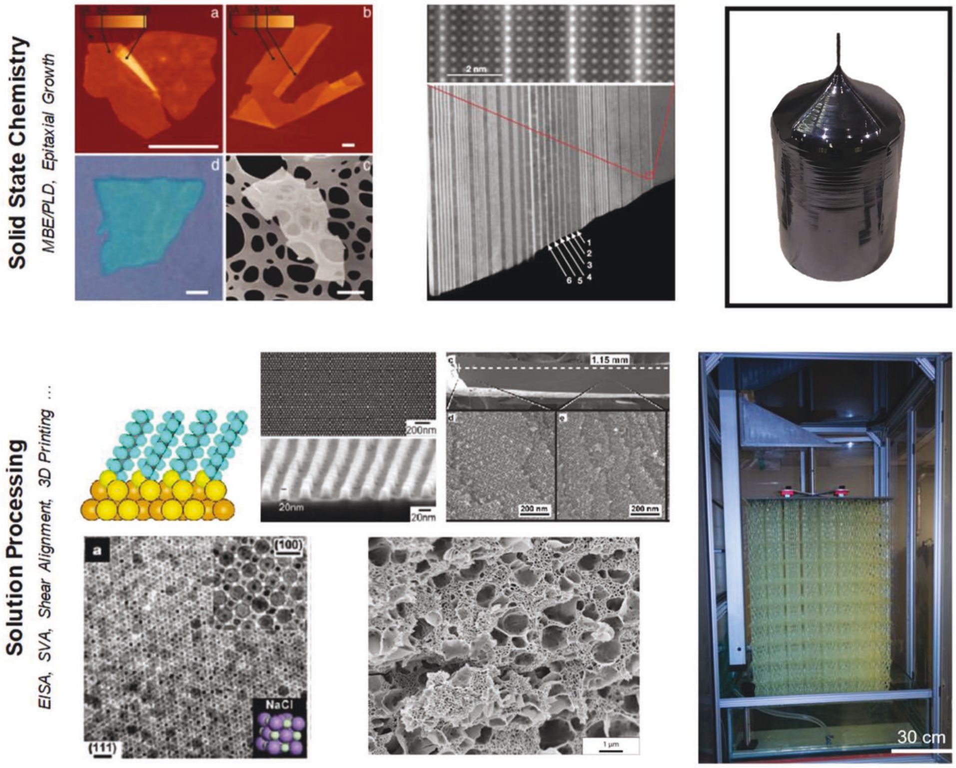

Figure 7. Comparison of typically available form factors in traditional solid-state synthesis (top) and soft-matter-based materials synthesis (bottom) methods. Top row from left to right: 2D single layer atomic crystallites of $NbSe_2$, graphite, BSCCO, and MoS_2; superlattice of single $LaTiO_3$ layers separated by $SrTiO_3$ of controlled thickness via PLD; and Czochralski (CZ) grown single crystal silicon ingot. Left: Adapted with permission.[287] Copyright 2005, National Academy of Sciences, USA. Middle: Adapted with permission.[288] Copyright 2002, Springer Nature. Right: Reused under CC0 terms. Middle row from left to right: depiction of a self-assembled monolayer (SAM) on a gold surface; Cr and Si dot/pillar thin film arrays made via lithography relying on the directed self-assembly of a diblock copolymer; triblock-terpolymer-derived gyroidal structured SiON ceramic consisting of a single macroscopic domain with coherent ordering from one side to the other. Left: Adapted with permission.[279] Copyright 2005, American Chemical Society. Middle: Adapted with permission.[290] Copyright 2008, American Association for the Advancement of Science. Right: Adapted with permission.[239] Copyright 2019, Wiley-VCH. Bottom row from left to right: NaCl-type binary colloidal superlattice; hierarchically porous polymer scaffold derived from the combination of spinodal decomposition and macromolecular assembly in a block copolymer containing system; 3D printing via high-area rapid printing (HARP). Left: Adapted with permission.[231] Copyright 2006, Springer Nature; Middle: Adapted with permission.[291] Copyright 2013, American Association for the Advancement of Science. Right: Adapted with permission.[292] Copyright 2019, The Authors, published by American Association for the Advancement of Science.

4.2. Thin Films

Above 2D materials is the very well-developed and well-studied form factor of thin film materials. Through the refinement of methods such as atomic layer deposition, MBE, and PLD, experimentalists have obtained precise control over parameters such as composition[288] and strain[304] in crystalline thin films.[305] This has contributed greatly to the study of 2D electron gases,[306] magnetic heterostructures,[307] and many other hybrid materials and emergent phenomena at interfaces.[308,309] The variety of techniques available for growing epitaxial heterostructures provides useful breadth in synthetic capabilities, but all suffer from limitations imposed by the stringent synthesis conditions required.

From a soft matter perspective, thin films are rich both in available mesostructures and material composition. It is worth noting that the thickness of thin films prepared from soft matter can range from the nanometer up to the micrometer scale, i.e., significantly thicker than the typical thin films in the physics or electronic device community. To fabricate thin films, using polymers as an example, homogeneous solutions of polymers in appropriate solvents are first prepared before casting onto a substrate. This requires the solvents to be able to dissolve the polymer and to have favorable wettability on the substrate. Dip coating involves immersing the substrate into the solution before gradually pulling out.[310] The more commonly practiced way is spin-coating, where solutions are spun off the substrate at a defined rotational speed, and a smooth thin film is derived after rapid solvent evaporation of the small amount of polymer solution that adheres to the substrate. Film thickness can be precisely controlled through varying the rotational speed and solution concentration (as well as secondary parameters such as acceleration).

Since the thin film is supported on a substrate, the energetics between the substrate surface and the thin-film materials can dictate their structures.[310] It is possible to engineer the surface energy to preferentially align the thin-film substructure, thereby growing mesoscale periodic structures epitaxially from the substrate.[308,311] Another route to prepare epitaxial thin films capitalizes on the guiding of topographical elements to direct the structures, termed graphoepitaxy.[237,312] On top of such 2D patterns, multilayers of mesostructures with well-defined orientations can be prepared,[313–315] allowing for use in nanofabrication and investigation of emergent properties related to quantum behavior.

The rapid evaporation of solvents during spin-coating often does not leave enough time for the thin-film structure to fully order. Post-spin-coating treatments can be employed to improve the periodic ordering through annealing, a concept borrowed from conventional hard inorganic material processing. Apart from thermal annealing, the solution-processable soft matter can be subjected to solvent vapor annealing,[317] which dramatically mobilizes the constituent species and shortens the annealing time to achieve high order. As is commonly applied in inorganic material processing, postdeposition annealing is one more knob to turn for structural manipulation without changing the material composition. By changing the solvent ratio, flow rate, annealing time, among other parameters, different morphologies can be obtained, e.g., form a single block copolymer[318] or other self-assembly enabling synthons and intrinsically 3D mesostructures can be fabricated without resorting to tedious multilayer deposition.[319]

4.3. Bulk Single Crystals

Beyond the thin film regime, advances in the synthesis of bulk single crystals have played an invaluable and enabling role for the discovery, study, and understanding of quantum materials. Whether guided by theoretical predictions or breaking new ground, the creativity and ability of crystal growers have led to many of the seminal discoveries in quantum materials.[17] Evaluation of the properties in any crystalline system is directly dependent on forging routes toward highly perfect bulk materials; defects, grain boundaries, and disorder in general obscure transport behavior and are often considered obstacles to be surmounted via synthetic optimization. In this pursuit, experimentalists can exercise an exquisite degree of control over atomic ordering via the wealth of techniques pioneered by semiconductor manufacturing: Czochralski (CZ) growth, float-zone methods, or Bridgman techniques among others.[320] This enables rational design of new quantum materials through such means as careful tuning of stoichiometry or precise doping.

Because of the inherently large defect density often present, obtaining bulk mesoscale single crystals through soft matter self-assembly is often a challenge. In anisotropic systems such as colloidal nanorods[321] or lamellar and hexagonal block-copolymer morphologies,[322,323] external fields such as shear have been used to great effect to produce macroscale alignment of mesoscopic domains. Much progress has also been made with isotropic particles in cubic colloidal crystals, where single crystals of multiple millimeters in size are now regularly reported.[267,324–326] Achieving the same level of long-range order has remained more elusive in networked block copolymer morphologies, but recent work has achieved gyroidal single crystals up to a centimeter in size. The preparation of single-domain periodic mesostructures on a macroscopic scale is tantamount to atomic single crystals (e.g., using float-zone or Czochralski method), and such samples are expected to enable sophisticated property measurements in a similar way.[239,327]

4.4. Additional Form Factors

Whether in the 2D, thin film, or bulk regime, from the foregoing discussion, it is apparent that soft matter methods have to progress toward the same level of order and structure control as already available in traditional solid-state techniques. While there is still much work to be done toward this end, soft-matter-enabled synthetic methods offer further benefits. Solution processing is not limited to thin film or monolithic bulk single crystals in the same way that traditional methods are, and thus soft matter enables a wealth of possible form factors. Colloidal superlattices, for instance, offer the opportunity to control symmetry/structure and composition across multiple length scales simultaneously.[231,268] Likewise, hierarchical materials can be made through such means as block copolymer self-assembly in tandem with spinodal decomposition[291] or phase inversion.[328] Furthermore, through additive manufacturing/3D printing methods such as continuous liquid interface printing (CLIP)[329] and high area rapid printing[292] (Figure 7), polymeric resins and polymer composites can be defined in arbitrary macroscopic shapes. This expansion in accessible form factors for quantum materials synthesis is but one of the contributions, which soft matter science could make to the field. In the next section, we explore a few more of these possibilities as examples of the promise of soft-matter-enabled quantum materials.

5. Future Perspectives

While previous sections have shown that there is a growing body of work bridging soft and hard condensed matter physics, there remains much to be explored in the use of soft matter for the synthesis and study of quantum materials. In this next section, we will explore a few paradigms for future work at this interface, organized under the following general thrusts: expanding structural control, mesoscale lattice engineering, tuning disorder and composition, considering increased interfaces, convergence in the thin film regime, and harnessing extreme conditions. A few vignettes will be used in each case as a way to showcase the opportunities for soft-matter-enabled quantum materials, but the general concepts underpinning each thrust can and should be extended beyond any single soft matter system or quantum material class.

5.1. Expanding Structural Control

As described, access to novel and often unique molecular and nano- to mesoscale architectures is one of the most salient strengths of soft-matter-enabled quantum materials; one of the biggest challenges facing their widespread impact is the inherent difficulty in creating well-ordered single-crystal-type materials with low defect densities. To fully realize the potential of soft matter in quantum material synthesis, structural control must be expanded and refined across the atomic, nanoscopic, and mesoscopic levels. The myriad of possible structures available through soft matter self-assembly is exemplified by the diversity in colloidal assembly. Thus far, only simple close packed assemblies of colloidal spheres have been translated into quantum materials. Synthetic advances in controlling shape and functionality of nanoparticles have led to a wide diversity in colloidal crystals, however, with many different packing geometries and lattice symmetries.[231,234,275,330] Translation of these architectures into quantum materials, e.g., via soft or hard templating would yield a vast array of novel materials to explore.

Future work should also address the challenge of achieving high degrees of ordering over macroscopic distances. Much work has been performed in order to align and perfect soft self-assembled structures, in thin film or bulk regimes as well as across different soft matter material classes.[232,237,239,331] At the same time, many studies have explored structure control via confined crystallization.[332–334] At the convergence of these two efforts is a long sought-after goal in the field of soft templating, i.e., the simultaneous control over soft and hard materials' ordering. As an example, it has been shown that single crystalline nanowires of various quantum materials can be grown via nanoporous templates.[335] As we described earlier, recent work has made available mesoporous gyroidal ceramic templates consisting of single, macroscopically coherent gyroid domains.[239] The combination of these techniques could yield a gyroidally structured superconductor, topological insulator, or artificial spin ice in which an atomic single crystalline material possesses a single orientation conserved throughout the bulk of a single crystal mesoscale structure. Accomplishment of such a hard single crystal in tandem with a soft single crystal, i.e., a "single crystal in a single crystal," would unlock the full investigation of emergent transport properties, angle-dependent behavior, or other unique effects of mesoscale ordering on quantum materials, but in general remains rather challenging to obtain.

5.2. Mesoscale Lattice Engineering

Soft matter, after all, is pliable and deformable, a characteristic that earned it its name. Soft matter also enables large-scale roll-to-roll manufacturing. In stark contrast with atomically crystalline hard materials, which typically render lattice engineering across substantial length scales challenging as a result of well-defined bond lengths and bond angles, soft matter offers substantive flexibility for lattice engineering of mesostructures. Therefore, we can identify a pathway where a palette of soft-matter templating materials can be adopted to direct the mesostructure formation of additives, which upon various chemical and physical transformation processes result in the desired relevant quantum materials (**Figure 8**). An exceptional feature here is that, through careful choice of materials and processing parameters, the mesostructure can undergo enormous deformations (e.g., as large as 75% changes in lattice constants) and symmetry breaking[338] unimaginable in hard matter, while still achieving highly crystalline atomic lattices at the local scale

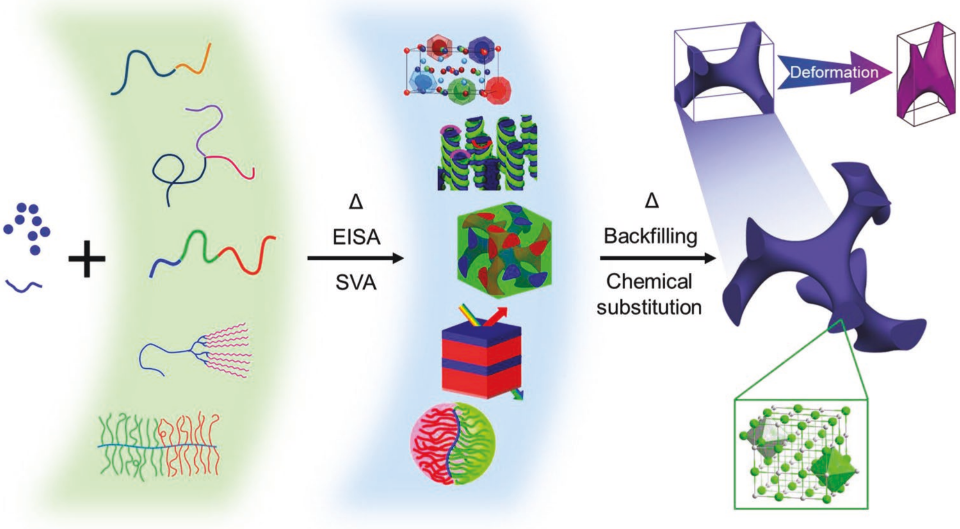

Figure 8. Schematic pathway from soft matter (using blocked polymer architectures as an example) to quantum materials. Block copolymers with tailor-made chain architectures and compositions (left, green background) can direct the mesostructure formation of additives that may be precursors of inorganic quantum materials (left, blue spheres) through solution-compatible processing steps, including evaporation-induced self-assembly (EISA) and solvent vapor annealing (SVA). Via a host of possible physical and chemical transformations, the resulting mesoscale lattices (middle, blue background, from top to bottom: Adapted with permission.[338] Copyright 2017, American Association for the Advancement of Science; Adapted with permission.[339] Copyright 2012, American Chemical Society; Own work; Adapted with permission.[336] Copyright 2012, American Chemical Society; Adapted with permission.[337] Copyright 2012, American Chemical society.) can be transformed into structures that confer mesoscale ordering on top of local crystalline atomic lattices of traditional quantum materials (right, using a gyroidal minority network in blue and a perovskite atomic lattice in the green inset as examples). Due to the soft nature of polymers, as well as their periodic lattice parameter dependence on the number of monomers along the chain, polymer-based mesoscale lattices are amenable to substantial variations of their lattice constants as well as deformations leading to symmetry breaking (upper right), both largely unavailable in atomic lattice engineering.

enabling the desired electronic, magnetic, optical, and phononic properties. Engineered soft matter structures or templates are also often transferrable to other arbitrary surfaces,[319,340] providing more material design choices, e.g., via backfilling and integration into devices, all thanks to their deformable nature.

5.3. Tuning Disorder and Composition

The myriad of achievable molecular and nano- to mesoscale structures and form factors offered by soft matter brings the notion of controlled disorder into the study of quantum materials. Long considered undesirable compared to crystalline order, structural disorder is often an intrinsic signature of materials derived from soft matter, from amorphous and semicrystalline polymers to nematic/smectic phases of liquid crystals. However, there has been growing evidence of the significance of disorder in the study of quantum materials.[2] Controlled disorder has manifested itself as an indispensable ingredient in unleashing intriguing quantum phenomena in Anderson topological insulators.[341] Dubbed amorphous quantum nanomaterials, glassy dye-encapsulating nanoparticles hold tremendous promise in photodynamic therapy, thanks to spin–orbit coupling.[342] In such glassy solids, atomic average distances of heavy atoms to the delocalized π-electron system of the encapsulated organic dye can be fine-tuned via substantial variations in composition, typically not achievable in crystalline solids without changing the crystal structure/nature, in turn providing new handles on tuning the strength of spin–orbit coupling.[342] Though the physical origins warrant further deeper understanding both theoretically and experimentally, these results clearly point in the direction of embracing the control of disorder as part of our toolbox for the design and application of soft-matter-enabled quantum materials.

Composition is a strong tuning knob for the design of quantum materials provided by soft matter. What is helpful in achieving compositional variability is that details of molecular structure, such as the organic ligands in metal–organic frameworks, the block chemistry of block copolymers, or the details of surfactant molecular structure, can often be neglected in understanding the observed mesoscale structure formation. Earlier examples, albeit from simulation work, demonstrated the possibility of designing various organic ligands that link metal centers to engineer the spin–orbit coupling effect for the creation of topological insulators.[179,181,182] Given a metal center's coordination chemistry and geometry, there is a wealth of molecular structures, and consequently compositions, that can be ingeniously fine-tuned by organic/material chemists to achieve practical real-life examples of topological materials. Such compositional variability does not come at the expense of structural integrity, since the self-assembly is a highly modular process where the interactions between soft matter building blocks are more forgiving than the exact bonding between atoms in typical inorganic quantum materials.

5.4. Considering Enhanced Interfaces

Nobel laureate Herbert Kroemer famously said "the interface is the device," in reference to devices realized with thin film semiconductors.[343] More than 20 years after he received his Nobel Prize in Physics, this statement still rings true. The importance of interfaces is a common thread in both mesoscale science and quantum materials.[24] In quantum materials, interfaces are often a key component in emergent quantum properties, whether they be used to realize a quantum behavior or have a direct effect on an already-emergent property.[208,344] Using self-assembly directed molecular and nano- to mesoscale structures and, in particular, micro- to mesoporous materials, means working with extremely high surface area. In the case of the BCP-directed gyroidal superconductors mentioned earlier, this high-surface area mesoporous material already is a quantum material. The substantial pore volume in 3D of such gyroidal superconductors can subsequently be backfilled with a host of other materials, e.g., in the form of insulators, semiconductors, metals, or another superconductor. This provides a route to 3D interpenetrating composite materials with large interfaces, and with exotic interface geometries, well beyond the common planar interfaces observed, e.g., in MBE- or PLD-derived materials. Considering the large field of self-assembly directed porous solids,[242,345,346] this is a vast and to date largely unexplored playground for the study of emergent quantum material properties.

As another example, it is known that $SrTiO_3$ and $LaAlO_3$, two insulators, exhibit superconductivity at their interface.[287] It has also been shown that a $SrTiO_3$ substrate with a FeSe film, already a superconductor with T_c of 9.4 K in the bulk, shows an extremely elevated T_c of above 100 K at monolayer thickness.[343] These unintuitive results of superconductivity at insulator interfaces, or drastically increased T_c over the bulk of a thin film at monolayer thickness, have led to interfacial superconductivity to be considered as a new frontier for the field.[346] Now regarding soft matter, it has been demonstrated that metal oxides can be structure-directed into high surface area, mesoporous materials with varying 2D and 3D geometries.[36,39] Combining these two concepts could give rise to superconductors with large area superconducting interfaces and with potentially more interesting geometries and properties than the traditionally planar interfacial superconductors. However, as previously mentioned, such a step forward would perhaps require a more precise level of control in structure direction than is typically achieved today.

It should be noted that, since mesoscale structures from soft matter self-assembly are typically the result of weak intermolecular forces, they are prone to thermal fluctuations leading to a variety of structural distortions not common to hard materials. While the study of such structural distortions is well developed in some areas of soft matter science, including liquid crystals where defect structures are readily discerned via the optical microscope, in other areas like BCP-self-assembly or directed-self-assembly (DSA) based soft crystal formation, where comprehensive understanding of distortions/defects involves some sort of electron microscopy, powerful experimental approaches for their quantitative assessment and understanding are still being developed.[240,348] It can be expected that the effects of such soft matter intrinsic structural distortions on quantum material properties will become a rich area of study, which may lead to novel physical phenomena with no analog in the hard matter field.

5.5. Convergence in the Thin Film Regime

Perhaps the most natural area for overlap between the soft matter and quantum material communities, and the area in which there is the richest potential for initial collaboration, the thin film regime is intimately familiar to those in both disciplines. Moreover, in both communities, there have been decades worth of inquiry and development of thin film fabrication and processing techniques. In traditional quantum material synthesis, many epitaxial growth and deposition methods exist which have been highly refined for the creation of thin film materials.[2,17] As shown by the examples in **Figure 9**, DSA of soft matter has often been demonstrated as a promising alternative method for bottom-up synthesis of nanoscopic features in thin films.[290,331,353–355] Cutting across both disciplines, providing routes to devices and motivation for research, are all the techniques associated with microlithography, nanofabrication, and semiconductor manufacturing.[356]

The application of soft matter for the synthesis of quantum materials in the thin film regime has already begun paying dividends, shown in earlier sections on superconductors,[114,115] topological materials,[155,165] multiferroics,[187,190] or spintronics.[79,198–200,210] However, future work in this area may seek to take full advantage of the established techniques and capabilities present in soft matter thin film formation for the creation of new quantum metamaterials or devices. To begin, soft matter is already ubiquitous in nanofabrication in the form of photoresists, liftoff layers, packaging materials, and many other forms. This ubiquity is born of utility; spin coating, direct patterning, thermal or solvent annealing, and etching all allow for an extremely high degree of control over thickness, areal coverage, material composition, and final film properties through relatively facile processing means. Through means such as coassembly and soft or hard templating, this processing ease may be transferred into the area of quantum materials which do not natively possess such desirable characteristics. This will enable more accessible and scalable routes toward quantum materials, a crucial step toward further research and widespread adoption. Further, the use of soft-matter self-assembly to define thin film quantum material structure likely will enable the observation of emergent metamaterial behavior. For example, interesting interfaces between superconducting oxides or topological insulators could be defined in 3D in interpenetrating networks, and fabricated into planar devices to make use of the plethora of available synthesis and measurement techniques. Multiferroic domains of crystalline materials could be defined on length scales from the atomic to the mesoscopic for the engineering of spin and charge properties, and then easily integrated into sensing platforms. Nanofabrication is an area in which both quantum materials and soft matter investigators have a large presence, and that is backed by a long history of device integration resulting in tremendous scientific and technological promise. In the area of thin films, a bridge from soft matter to quantum materials has already been created, and invites heavier traffic.

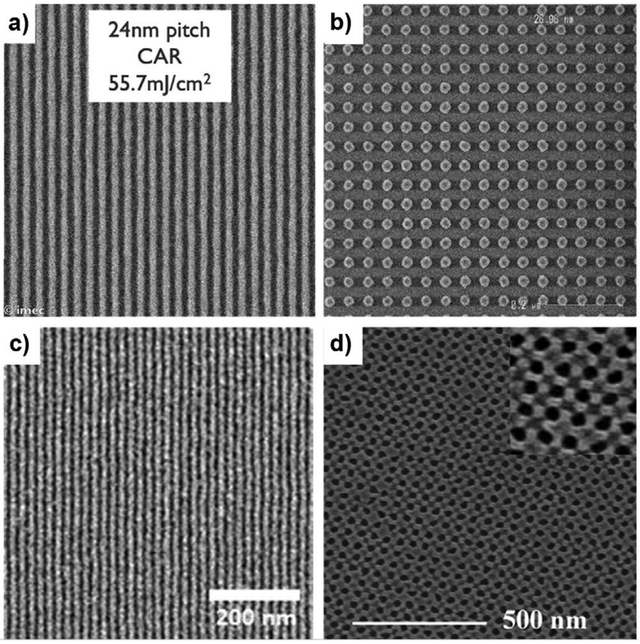

Figure 9. Comparison of conventional lithographically patterned structures (top) with those obtained via soft matter self-assembly (bottom). a) Demonstration of 24 nm pitch lines via single exposure EUV lithography. Adapted with permission.[349] Copyright 2020, Interuniveristy Microelectronics Centre. b) 50 nm pitch pillars fabricated via EUV lithography. Adapted with permission.[350] Copyright 2019, Society of Photo-Optical Instrumentation Engineers. c) 20 nm pitch lines fabricated via directed self-assembly of a silicon containing block copolymer. Adapted with permission.[351] Copyright 2016, American Chemical Society. d) 50 nm pitch pores fabricated in silicon via self-assembly and selective etching of a block copolymer. Adapted with permission.[352] Copyright 2008, American Association for the Advancement of Science.

5.6. Harnessing Extreme Conditions

Extreme conditions are common in quantum material synthesis, from high temperatures in laser annealing[48] to ultra-high pressures.[50,51] Such methods may at first glance seem to preclude the use of soft materials. As it turns out, however, through careful choice of processing parameters, the instability of soft matter under these conditions can often be overcome. For instance, by setting the time scale of transient laser annealing processes below that of the chemical reactions governing soft materials decomposition, one can “outrun” their oxidative degradation kinetics.[37,357,358] Even though at first sight this sounds impossible, using transient laser annealing, this concept has already enabled thin film porous polymeric templates to structure direct liquid silicon, with a melting temperature well above 1000 °C, into 3D porous silicon nanostructures.[34] Moreover, extreme processing conditions have uncovered previously unknown material phases or may stabilize previously inaccessible metastable phases.[359] These early and encouraging experimental results open up a whole new range of nonequilibrium and extreme processing conditions that may become available to generate soft-matter-enabled quantum materials. This research direction also poses a number of interesting questions. For example, what is the ultimate tolerance of soft matter under these harsh conditions and how can that tolerance be extended? How can we identify additional extreme conditions appropriate for streamlining material discovery and application? Given more questions asked than answered, this new paradigm encourages creative thinking out of the box.

6. Conclusions

While the fields of soft matter and quantum materials in the minds of many have so far remained largely separate, we have presented here a review of work which begins to bridge this divide. This body of research is growing at an accelerating pace, and hints at the great potential of the emerging field of soft-matter-enabled quantum materials. By describing the landscapes of both of these original fields, and areas where they start to overlap, we have elucidated some of the potential for scientific discovery and innovation. But clearly much remains untapped. The field of soft matter and its self-assembly is already highly developed, and advances in our understanding of associated phenomena have already paid enormous dividends, from paint dispersions to cosmetic products, to functional devices, such as organic light emitting diodes and lithium-ion polymer batteries,[360,361] to increased stability of novel vaccines via self-assembly processes.[362,363] Advances in the field of quantum materials science could revolutionize, e.g., the information science sector, changing how we communicate and compute,[2,3] how we move people and goods from point A to B, or how we interpret imaging data in health care. If the acquired knowledge in the field of soft matter is fully brought to bear on the preparation and study of novel quantum materials, the resulting expansion of scope beyond the more traditional pathways to this exciting materials class would open up an enormous space for exploration and innovation.

As previously stated, the importance of interfaces is common to both soft matter and quantum materials. Interfaces modulate the thermodynamics of self-assembly, the appearance of topological insulators, and the transition temperature of superconductors. The convergence of the two fields of "soft" and "hard" condensed matter science provides tremendous opportunities and substantial challenges. With much work to be done in order to address such challenges and realize the full potential of soft matter in quantum material design, synthesis, science, and engineering, we hope that this review will inspire researchers to take interest in working "across the aisle." It is only by reaching through this interface that we may fully realize the promise of soft-matter-enabled quantum materials.

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Note: The copyright attribution for Figure 4a,c was clarified as both panels being attributed to ref. [33] on February 2, 2023, after initial publication online.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

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