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Perspective

Topological Dynamic Matter

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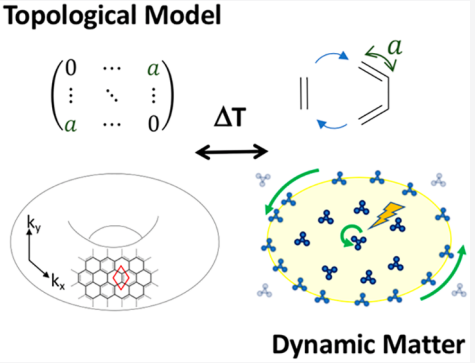
ABSTRACT: The principles of topology in condensed matter physics have expanded to areas such as photonics, acoustics, electronics, and mechanics. Their extension to dynamic (soft) matter could enable the control and design of topological thermodynamic (micro)states and nonreciprocal dynamics, potentially leading to paradigmatic applications in molecular and thermal waveguiding, logics, and energy management. This Perspective explores distinct topological concepts for dynamic matter and prospective function. Topological tools are exemplified and discussed for the study of nonlocal order parameters or invariants in dynamic molecular matter, toward the engineering of assemblies, reactions, and system chemistry with unconventional global properties—a scope which has the potential to push the frontiers of physical chemistry and transform chemical topology from form to function.



Topology and corresponding order parameters have substantially contributed to the understanding of complex systems for centuries.[1−3] In the context of (bio)chemistry, topology has been employed to analyze the shape or form of polymers, (supra)molecules, proteins, and chiral liquid crystals.[4−10] Conversely, our perception of applied topology in physics has been shifting from the description of form to a property of waves and particles in bands[11,12] and intrinsic quantum order,[13] with extraordinary functional implications. This has led to a paradigm shift in condensed matter which has now extended to quantum computing, ultracold atom science, photonics, phononics, and mechanics. Protection against disorder, nonreciprocity, and waveguiding are among the extravagant topological band properties which have been demonstrated at boundaries between materials, and their realization at finite temperatures holds great promise for the (bio)molecular and soft matter fields.[14] In this Perspective, we outline diverse topological concepts related to dynamic molecular matter. The background section introduces notions of band topology familiar to condensed matter physics (Scheme 1), along with representative applications of noninteracting symmetry-protected topological states (Figure 1). The second section offers experimental examples related to topological defects and reaction topology, at transitions between topological configurations (Figure 2). The third and fourth sections discuss micro- and macrostates from effective band topology models, relevant to thermal (Figure 3) and matter (Figure 4) transport. This panorama may stimulate the exploration of broadly defined topological dynamic matter inspired by effective models in quantum matter, that is, the exploration of molecular, biological and colloidal active matter expressing functional topological nonlocal properties.

Background. In the past, particles or waves in solids were studied locally, departing from the connectivity of the atoms in a (minimal or primitive) unit cell (Scheme 1, red). Such modeling undermined the role of the connectivity and symmetry at the boundaries of real materials, which were once thought unnecessary for the description of their bulk properties. Topological tools enable the study of nonlocal order, connectivity, and symmetries of matter. The mathematical branch of topology develops algebraic or differential tools to study shapes: symmetric or compact spaces such as rings, tori, and matrices (Scheme 1a−c).[3] In such compact spaces, an order parameter (topological invariant) can be defined and associated with the finite material or system with boundaries (Scheme 1d−f).[12] Nowadays, applied topology employs topological models and invariants to classify unconventional physical properties of materials both in time and in space. In the archetypical electronic band structure example leading to the 1985 and 2016 Nobel Prize in physics (the integer quantum Hall effect, QHE[18]), the local atom connectivity in the crystalline unit cell alone could not explain the quantization and robustness of a measured quantity (the von Klitzing constant $h{\cdot}e^{-2}$) in the presence of a magnetic field. Instead, topological tools for characterizing the (Berry) connectivity between electron's momenta[19−21] helped explain the conductivity along the boundary of the finite solid (the Hall conductivity): The

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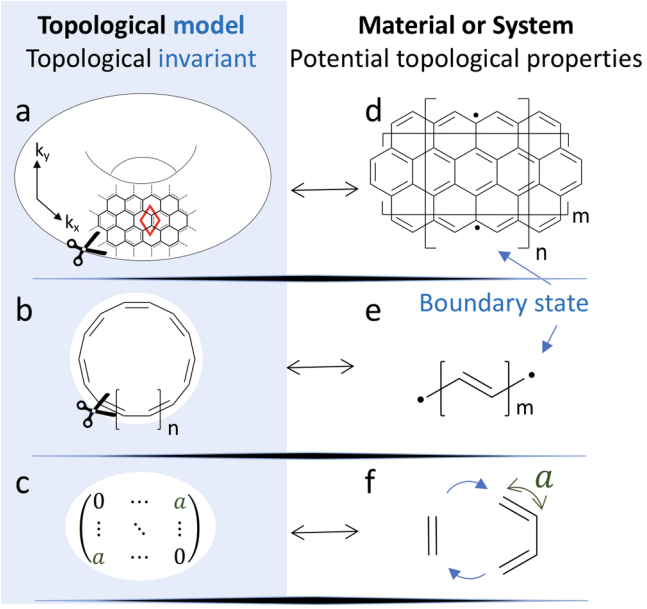
Scheme 1. Applied Topology in Physical Chemistry: Properties of Matter beyond Local Connectivity[a]



[a]Topological tools enable the nonlocal study of properties and connectivity in matter, *i.e.*, beyond a local unit cell (in red). In differential or algebraic topology, usually a smooth (eigen)function in a discrete material or system such as (quasi)momentum ($k_{x,y}$), is modelled within a compact space, *e.g.*, (a) on a torus, (b) on a circle or, (c) via its connectivity matrix.[15] The corresponding finite material, such as (d) a graphene nanoribbon cut from graphene,[16] (e) a diradical polyacetylene chain,[17] or even a (f) Diels–Alder reaction[15] can be described as a phase in the model and thus may reflect topological properties and boundary (edge) states. Topological models could therefore anticipate multiradical ground states, reaction yields and potentially, thermodynamic states.

quantization and robustness of the von Klitzing constant was thus revealed when the Hall conductivity was found proportional to an integer, an order parameter, or nontrivial topological band invariant related to the Berry connectivity via the Chern–Gauss–Bonnet theorem. One mathematical interpretation of topological triviality for vectors along a path (a vector bundle) is their reducibility to the trivial point. In the context of (molecular) materials, the trivial point may represent the single atom (or molecule) or the uniform atomic (or molecular) background. At such "atomic limit",[22,23] it is safe to assume that a material or solid can be simplified as a local unit cell of its underlying components. In contrast, a nontrivial topology epitomizes nonlocality. In the QHE, nontriviality is expressed in the form of metallic-like boundary (edge) states, and sometimes an insulating bulk, whereby the terms topological insulator and topological edge states originate. The strategy of engineering topological edge states in condensed matter physics has now extended to photonics[24,25] and circuits,[26] beyond correlated quantum materials.[11]

Because of nonlocality, microscopic states of nontrivial topological matter may express important properties of band topology such as protection against disorder, nonreciprocity, and waveguiding, to mention a few (Figure 1). Take for instance the topological photonic state of a coupled ring laser microarray[27] mimicking the Su–Schrieffer–Heeger (SSH) model,[17] which will be described in the next sections. Lasing occurs from the single-mode edge state as a consequence of the

Topology develops algebraic or differential tools to study shapes: symmetric or compact spaces such as rings, tori, and matrices. *Applied topology* employs topological tools, models, and invariants to classify the physical properties of matter.

array topology; in this case, the boundary state "soliton" is localized near the central ring (Figure 1a, top panel). Single-mode lasing is robust against an induced defect, introduced by a polymer layer (white square). In contrast, a trivial microlaser array emits as a broadband laser, and defects affect its spectral energy. Topological band insulators are not only restricted to the quantum realm[28] but can be defined mechanically[29−31] and acoustically.[32−34] The *portmanteau* topological insulator effect, the quantum spin Hall effect (QSHE),[35] is closely related to the integer QHE except for two aspects. First, instead of an externally applied magnetic field, an equivalent (internal) excitation is present. This is reproduced in the so-called Chern insulator (CI)[16] model. Second, the excitation is symmetric in time, meaning that two, opposite spin 1/2 channels counter-propagate (Kramers pairs). Spin-Chern insulator (spin-CI) and related models include these effects.[35−37] Spin 1/2 channels can be emulated classically,[29] by engineering double Dirac cones with different polarizations for instance.[33] With the appropriate excitation and edge channels, a "thermal diode" at a frequency window ($\Delta\omega$) can be, in principle, devised from a three-input terminal as in Figure 1b, right.[38] Such boundary (edge) states evolve at interfaces between effective topological phases (see next sections). Interfaces joining different topological domains can be arbitrarily (re)configured into edge state waveguides, for logics or storage applications. For example, the simulated steel-pillar lattice in Figure 1c[39] can be distorted from a trivial to a nontrivial topology for the prospect of engineering switchable acoustic waveguides.[33,39,40] Following the success of photonics and acoustics, the topological characterization of thermodynamic materials is expected to expand the frontiers of physical chemistry. Formal equivalences between the aforementioned topological band models and open systems at the thermodynamic limit are the subject of intense research.[41−44] Together with band topology, general topological notions are an important departure point for the exploration of nonlocal order in molecular systems and soft matter. For instance, in the study of topological defects, molecular orientations define vectors along a path for dynamic differential topology studies. In topological reaction models, atomic displacements can enforce symmetries familiar to algebraic analysis. These combined perspectives set the scene for dynamical topological states of matter at finite temperature.

Configurational Topology. A prospective field of study in dynamic topology beyond band theory is related to "configurational" topological order parameters within a dynamic system. An example of one order parameter is the genus (g), an integer ($\mathbb{Z}$), which counts the number of holes in a compact object. It can be related to the Gauss–Bonnet theorem (Figure 2) and posed for common real-space topologies pertinent to supramolecular chemistry: A donut has a $g = 1$ and a plate has $g = 0$. Consider a supramolecular rotor in Figure 1a which can be

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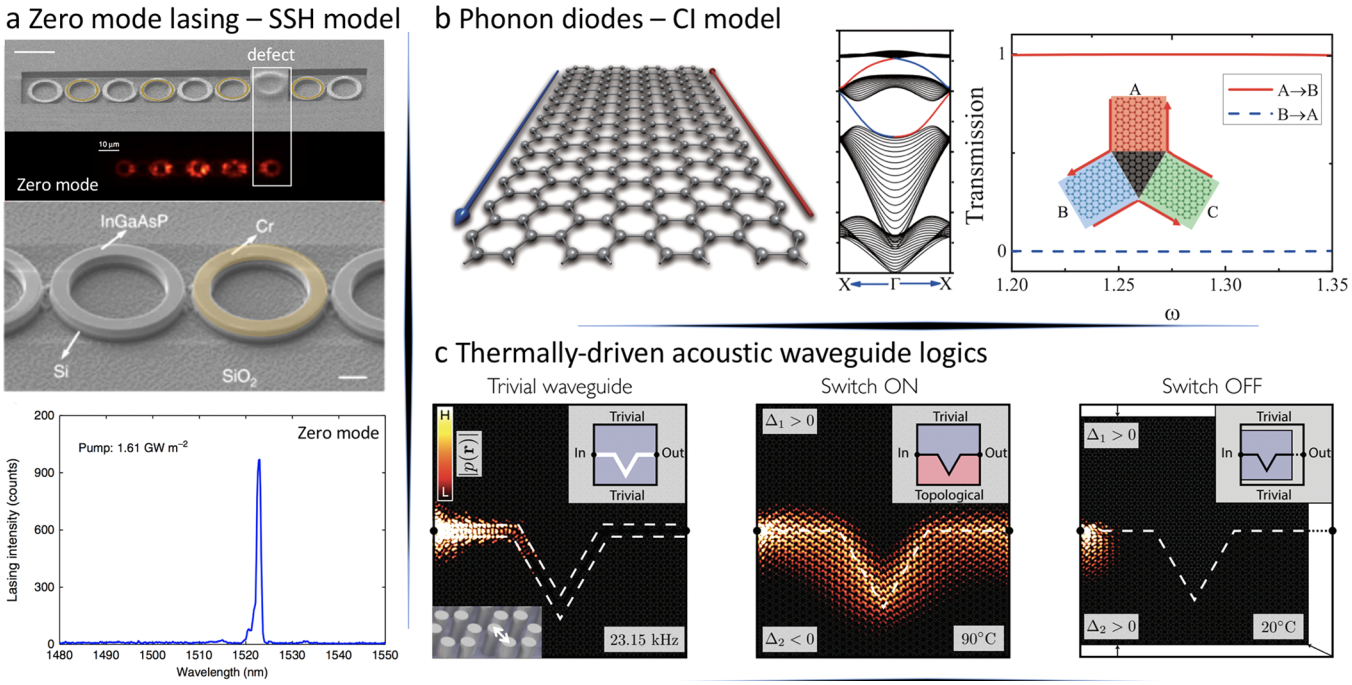

Figure 1. Examples of photonic and phononic topological function. (a) Robustness,[27] (b) nonreciprocity,[38] and (c) waveguiding[39] are hallmark boundary functionalities enabled by the control and engineering of topological matter. (a) A single-boundary state ("zero mode"), localized at the center of a microlaser SSH array, is robust against defects (square) in the array. Adapted from ref 27. Copyright CC BY 4.0. (b) Boundary (edge) phonon channels along a two-dimensional material can mimic a Chern insulator (CI) model in the presence of an excitation, toward efficient thermal diodes.[38] Adapted with permission from ref 38. Copyright 2017 American Physical Society. (c) An acoustic crystal simulated from steel pillars (inset) can be distorted between its trivial and nontrivial topology by means of thermal regulation of acoustic gaps (Δ_1 and Δ_2) to define a thermally driven sound switch[39] along its boundary (white dashed line). Adapted with permission from ref 39. Copyright 2018 American Physical Society.

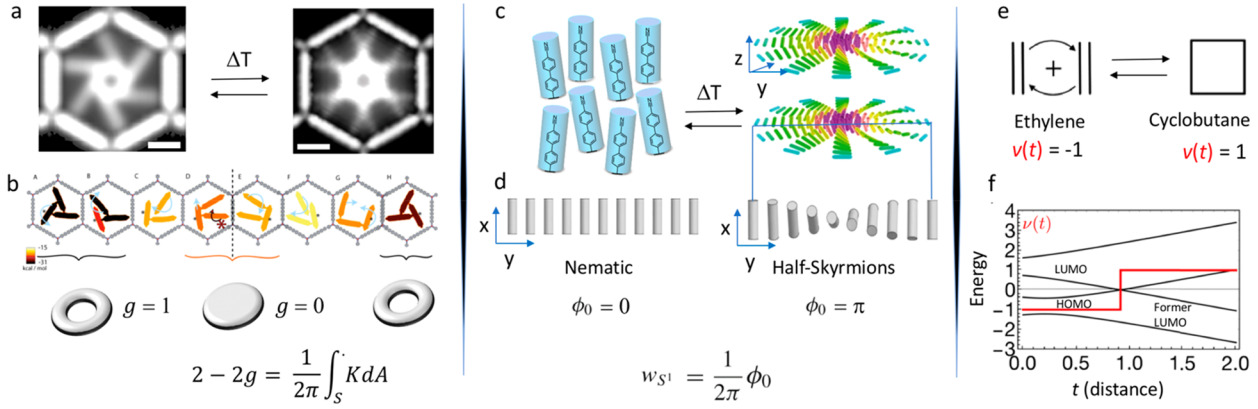

Figure 2. Topological classification of molecular configurations. (a) Supramolecules rotate while conserving their propeller shape below 87 K. At higher temperatures (b), the propeller intermittently breaks to allow a chiral inversion of the propeller. The propeller can be characterized by its genus (g), a topological invariant[45] related to the Gauss–Bonnet formula: an area (A) integral of the Gaussian curvature (K) for solids without boundaries. (c) The LC-1289 liquid crystal mixture can transition from a planar nematic to a half-Skyrmion (meson) phase.[48] The half-Skyrmion phase can be characterized by (d) its winding number (w_{S^1}), a topological invariant. The winding number measures in this case the number of π twists.[50] Adapted with permission from ref 48. Copyright 2017 Springer Nature. (e) A chemical reaction may also undergo a topological phase transition, shown by the reaction coordinate (t) in panel f. The transition is characterized by a topological invariant $\nu(t)$[15] related to the connectivity (site-basis tight binding Hamiltonian) matrix. Adapted with permission from ref 15. Copyright 2020 American Physical Society. (f) Noninteracting highest occupied molecular orbital (HOMO) and lowest occupied molecular orbital (LUMO) eigenvalues along the reaction coordinate (t) and topological invariant (red curve).

realized on a silver surface[45,46] following the deposition, self-assembly, and confinement of dicyanosexiphenylene molecular linkers with cobalt atoms under ultrahigh vacuum conditions. Because of the boundary conditions (confinement in the hexagonal pore of the lattice), three molecules define a propeller with $g = 1$ (Figure 2b). The propeller conserves its topology when rotating inside the hexagonal pore during the observation time scale (hours) at temperatures up to 87 K. The simulated dynamical pattern in Figure 2a results from two long-lived conformations of the propeller differing by 60° rotational symmetry. At higher temperatures, the topology of the propeller breaks via one of the mechanisms depicted in Figure 2b and an inversion of chirality is possible, yielding a markedly different dynamical pattern in Figure 2a, right-hand side. Knots and twists are also nonlocal order parameters and can be manipulated between boundaries for distinctive molecular self-assem-

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bly.[10,47−50] Consider the transition between a planar nematic phase and half-Skyrmion phase of the liquid crystal in Figure 2c, which has been predicted to occur when heating of a planar nematic phase or quenching of a high-temperature isotropic phase[51,52] and has been observed in ∼250 nm thin layers.[48] The transition occurs as the molecular wall or chain twist by an angle (ϕ) of π degrees. The twist is a nonlocal order parameter, and a winding number can be formally defined[10] as a topological invariant. The winding number describes the half-Skyrmion (or meson) for systems which can assembly into square unit cells (Figure 2c,d). Such half-Skyrmions and their full $\phi = 2\pi$ counterparts, Skyrmions, are favored when forcing boundary conditions (e.g., between glass slides) and can be manipulated by electric fields and optical tweezers.[47−50] The robustness of half-Skyrmions is evidenced when considering their planar (or vertical) uniform limit: Once self-assembled, half-Skyrmions would necessarily require breaking supramolecular connectivity to untwist them. As an example, consider a one-dimensional $\phi = 2\pi$ chain topology, inside a uniform planar nematic liquid crystal, that is, a chain featuring twice the twist shown in Figure 2d. The $\phi = 2\pi$ chain is trivially winded and can be unwound back to $\phi = 0$ (or $\phi = 4\pi$) by translating the start of the chain toward its end and vice versa (similar to holding a belt with both hands and exchanging hands). This is not the case for $\phi = \pi, 3\pi, 5\pi$, and so on, which would necessarily require a rotation, thereby tearing the uniform, vertical boundary in order to set $\phi = 0$. Thus, an associated order parameter for twisted chains is, in principle, not an integer ($\mathbb{Z}$), but mod 2 integer ($\mathbb{Z}/2\mathbb{Z}$ or equivalently $\mathbb{Z}_2$). Formally, mathematical homological mappings are often required to distinguish equivalent trivial topologies from nontrivial ones.[53] In homology, the order parameter is first mapped to a mathematically symmetric space such as the unit circle ($\mathbb{S}^1$), which avoids ambiguity in the definition and calculation of a topological invariant (cf. $w_{\mathbb{S}^1}$ in Figure 2d). Thus, algebraic groups are part of the topological space toolbox, as much as a circle or torus. As such, it is often sufficient to look at the algebraic symmetries of an associated matrix to define a topological invariant. Such a symmetry-protected, algebraic "topological periodic table"[54−56] has been described extensively and often arises when Hamiltonians or density matrices are employed to describe the dynamics of a system, as exemplified in the next sections.

Reaction topological models may contribute to the chemical kinetics of self-assemby, folding, and reactivity.

In the previous examples, we mentioned how different topological phases can evolve with temperature. What happens at the transition boundary between two topologies, configurational or otherwise? In band topology, the energy gap closes to allow for a topological band invariant change, resulting in boundary states (see next section). In topological reaction models,[15] exceptional points can occur at transition boundaries, and might indicate nonadiabaticity. For instance, it has been recently proposed via a tight-banding formalism[15] that the electronic structures of ethylene and cyclobutane have different topologies (Figure 2e). When changing the distance (t) along the reaction coordinate between them, so as to force a reaction, the reaction's topological invariant changes because of the (interacting or noninteracting) "crossing" between the highest occupied molecular orbital (HOMO) and unoccupied LUMO in Figure 2f. The reaction between ethylenes to form cyclobutane is thermally unfavored, proceeding rather in the photoexcited state, following Woodward−Hoffmann frontier orbital analysis. This is reflected in the change of topological invariant during the reaction in Figure 2f, further substantiating its nonadiabatic character. A related prospect regards topological folding[9] at the interface between configurational topology of complex soft matter systems and topological symmetric spaces. When a polymer is confined to a symmetric space, a new "folded" real-space topology may evolve at finite temperatures.[57,58] The ensuing configurational topology could be driven by both the symmetries of the target confinement in algebraic space and the intrinsic topological classification of the polymer connectivity.[4] Algebraic representations[15,57] of such folding processes could be suitable for quantum folding algorithms.[59] In this manner, reaction topological models may contribute to the chemical kinetics of self-assembly, folding, and reactivity.

Rovibrational and Phonon Topology. Rovibrational states defined by quantum numbers, similarly to electronic atomic states, are a consequence of the apparent trivial connectivity between momentum degrees of freedom in a molecule or molecular architecture. For nontrivial connections in time and energy, topological invariants may acquire the significance of quantum numbers.[60] Such topological band classification is different than in the previous section, in that configurational topology does not explicitly couple[61] to energy or momentum degrees of freedom. A quantum number precedes momentum conservation and thus defines (quasi)particle excitations, opening intriguing perspectives in energy transport and energy conversion. Thus, a common departure point to study band theory from a topological point of view is to consider one *portmanteau* matrix class[54,55] corresponding to the connectivity (energy function or Hamiltonian) matrix for Fermions or bosons under periodic boundary conditions. Hamiltonians of systems which present chiral (or sublattice) and pair (or particle-hole) symmetries such as the SSH model[17] can be described by a matrix class such as a Cartan class.[54,55] The topology and eigenvalues in such classes are simplified in terms of Pauli matrices (Supporting Information). Through several changes of variables, the phonon (bosonic) equivalent of the (fermionic) SSH model may also be expressed in a Cartan class space.[29] An alternative, intuitive approach to understand the topology of the phonon SSH model[62,63] is to consider the vibrational mode of the SSH model at the center of the unit cell (Brillouin zone wavevector $k = 0$) and at the edge of it (wavevector $k = \pi$). Solving the (SSH) phonon model for a periodic chain consisting of different spring constants (Figure 3a),[62−64] one finds two limiting cases. In the first case, the stronger spring constant ($+\delta$) lies at the center of the unit cell and the chain can be considered as approximately independent diatomic molecules. The top phonon band hosts an optical mode, characterized by an out-of-phase (oop) stretch, with atoms moving at opposite directions from each other (Figure 3b, left). In the second case, the top band features a hybrid behavior (Figure 3b, right). At $k = \pi$, the mode transforms to an in-phase (ip) displacement of neighboring atoms. Only at $k = 0$ is the oop stretch recovered (Figure 3b, right). In other words, the top band is neither fully optical nor fully acoustic. Instead, every diatomic pair alternates between oop and ip oscillations within a band, a phenomenon colloquially termed band inversion. A

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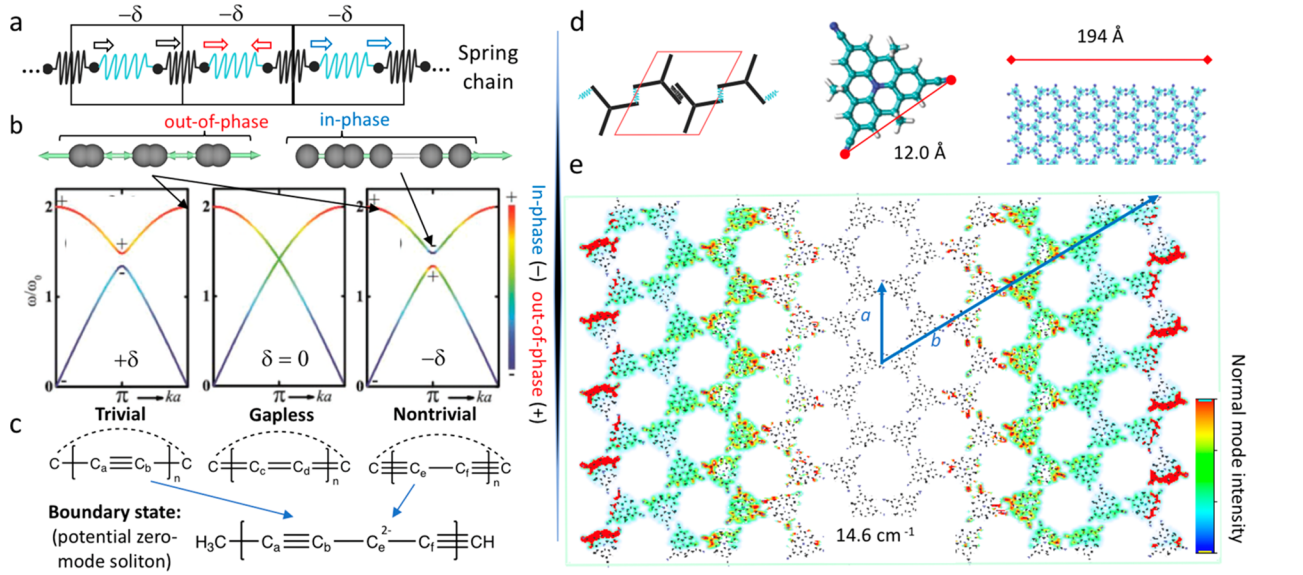

Figure 3. Topology of molecular vibrations. (a) The phonon SSH model. When the spring constant at the unit cell center is weaker by $-\delta$ (cyan spring), the optical band (top in the frequency ω band diagram) at the edge of the Brillouin zone transitions from the trivial case (b) out-of-phase (top band left, red color) vibration, to both in-phase and out-of-phase (top band right, blue color) vibrations.[62] Adapted with permission from ref 62. Copyright 2020 John Wiley and Sons. (c) Such model expresses topologically protected edge states when a boundary is created by fixing the stronger springs to a wall[62] and can be hypothesized in an atomic chain as follows: Stitching the trivial and the nontrivial topologies of carbyne and assuming stability, would yield a floppy boundary zero-energy mode (or soliton) at carbon C_e. (d) A realization of boundary vibrational states could involve an array of 1D SSH "supramolecular" chains (left), joining to form a supramolecular ribbon (right). (e) Molecular simulations show[79] that localized boundary states evolve in this manner, toward phononic supramolecular (spin-)Chern insulators.

dephasing along the length of a chain can be represented as a vector twisting in the complex Bloch circle, in a manner similar to the 1D twist chain, and it amounts to $\phi = \pi$. A related topological invariant is the Zak phase,[65] a particular case of the Berry geometrical phase in a single 1D band. The band structure in Figure 3b adapted from ref 62 shows with color the dephasing along the Brillouin zone. The nontrivial case is shown in the right-hand side of Figure 3b, whereas the trivial case is shown in the left-hand side. The intermediate case for the 1D phonon chain, where all spring constants are equal ($\delta = 0$) and the optical and acoustic bands meet, is also depicted. In appearance, the difference between the trivial and the nontrivial situation is defined by the unit cell choice. Take the hypothetical example of a trivial and nontrivial phonon topology for the periodic carbyne polymer in Figure 3c. The two forms appear misleadingly identical. However, the different unit cells portray distinct boundaries for the open system, which are better illustrated when connecting one unit cell to another, potentially defining a floppy boundary state or "zero mode" vibrational soliton (assuming the electronic structure remains unchanged, Figure 3c bottom). Such abstractions play an instrumental role in the design of topological boundary states in real materials, as discussed next.

In the previous sections, we learned that band topology studies trivial and nontrivial phases in periodic materials, whereby new boundary states may emerge at the interface between different topological phases. This phenomenon is known as the bulk-boundary correspondence (in Hermitian systems[66,67]) and may be accompanied by changes in the bulk density of states (for non-Hermitian systems[56,68−73]). A topological edge, corner,[74] or arc state[75] enables the extravagant functionality sought when engineering topological materials. Boundary states extend over macroscopic crystal lengths and feature finite bandwidths in the case of zero-energy mode flat-bands (in the analogue of the SSH model) or enhanced bandwidths with group velocities $d\omega/dk > 0$ in the analogues of the CI model. Predicting topological states in crystalline materials[22,23,76−78] at finite temperatures is a challenging endeavor (see below). Therefore, topological boundary states are usually characterized and demonstrated *ad hoc* in diverse functional settings, from diodes and logics to telecommunications and lasing (*cf.* the background section above). Two-dimensional supramolecular ribbons are ideal testbeds for inferring and probing topological properties in supramolecular matter.[79] A cyanotriangulane molecule at Au(111) interfaces self-assembles into a chiral lattice with a unit cell reminiscent of a SSH phonon model (Figure 3d). Simulations show that upon lifting periodicity to form supramolecular nanoribbons, new edge phonon bands evolve at the vacuum−ribbon boundary. The boundary eigenstates are found between gaps in the bulk band structure. The real-space projection depicts articulated boundary states, exemplified by the mode at 14.6 cm^{-1} in Figure 3e. The functional properties of the boundary states are probed by a sinusoidal excitation of a single supramolecular bond, resulting in unidirectional energy transport of the excitation. Interestingly, the excitation can propagate unidirectionally under low-dissipation Langevin simulation environments at 30 K, suggesting robust nonreciprocal transport arising from protected topological states at finite temperatures. As introduced above, topological invariants associated with topological edge phonons are well-known for algebraic, symmetry-enforced mechanical toy models[30]—yet mandate careful analysis in atomistic materials.[80] Moreover, topological tools at finite temperature formally differ from the topological invariants we have introduced, such that boundary states at finite temperatures cannot be formally inferred in the aforementioned "Hermitian" SSH model. Once lattice-temperature, dissipations, and fluctuations are considered, symmetries break down, either

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because of new (non-Hermitian) terms in the Hamiltonian or because of degrees of freedom acquiring different symmetries.[43,81] Topological states in open and thermal systems are currently the subject of intense theoretical research,[41−44] along with related boundary effects such as skin states.[70,71] Experiments on interfacial[82−84] and related model[85] molecular systems are expected to complement theoretical research on thermal (non)equilibrium dynamic topological matter, similarly to cold atom research.[81] Moreover, configurational topology representations of polymers and biopolymers could serve as input for archetypical topological phononic Hamiltonians.[44,54,55] For example, one simplified approach would depart by analyzing proteins into separate circular (periodic) chain segments and expressing the coupled harmonic wave equations within each segment via a first order differential equation, so as to define topological invariants following SSH mechanical models.[29] Each segment could be defined by the protein's geometrical circuit topology.[9] Algorithms could then find connections between topological nontrivial and trivial protein segments, where robust topological zero modes would be localized. The presence of zero modes between protein fragments has been hypothesized to aid in microtubule polymerization dynamics.[44] This view can be generalized to interpretations whereby edge modes play a role in protein activation, catalysis, and related phenomena.

Molecular systems are expected to complement theoretical research on thermal (non)-equilibrium dynamic topological matter.

Thermodynamic Topology. Thus far, the configurational topology of thermodynamic phases, and phonon topology potentially pertienent to thermodynamic microstates in lattices was defined, but can topology play a role in (out-of-equilibrium) thermodynamic macroscopic states and open molecular systems? Anisotropic thermodynamic states with salient nonlocal symmetries could lead to topologically articulated, robust molecular transport properties. Yet the thermodynamic aspects of real-world materials inevitably force nonlinear and chaotic dynamics, making experimental design and validation of "topological thermodynamic states" a grand challenge. Over the past few years, stochastic granular models and simulations[86−88] have stepped in to explore this exciting possibility, and stimulated the study of elasticity and hydrodynamics from a topological band standpoint. Recent examples are guided mostly by principles derived from CI models,[16] driven via, *e.g.*, internal excitations, and underlying nonequilibrium conditions. Consider for instance driving molecular rotors unidirectionally with light (Figure 4a), as means to break time-reversal symmetry.[89] A vortex boundary state evolves in such systems at the light-on, light-off interface (arrows around green periphery in Figure 4a). For the purpose of this Perspective and assuming small deviations from equilibrium, thermodynamic boundary states could be rationalized by the coupling between translational and rotational degrees of freedom at interfaces, and a tentative rotranslational free energy deviation (ΔF_{rot}, Figure 4b,c) close to the equilibrium state (gray models, Figure 4b,c). In ref 89, molecular dynamic simulations predict a localized boundary state, as depicted by the time-averaged velocities of the center of mass of the rotors, red triangles in Figure 4d. The underlying model can be described by the hydrodynamics of spin−vorticity coupling, via the constant (Γ), which represents the tendency of rotors to drag other rotors along with it. Solving for the vorticity (ω, the curl $\nabla\times$ of the flow velocity), the authors demonstrate that boundary states localize at interfaces irrespective of their shape and defects. To study the topology of the hydrodynamic model, the authors show that the hydrodynamic operator[86,90] $H^2 = (-\nabla^2 + \lambda^{-2})$ with λ^{-2} proportional to the angular or linear damping times the identity matrix, can be mapped in terms of the 2D Dirac band model Hamiltonian (Figure 4e,f). An effective "Dirac mass" $m \sim \lambda^{-1}$ in a Dirac Hamiltonian leads to

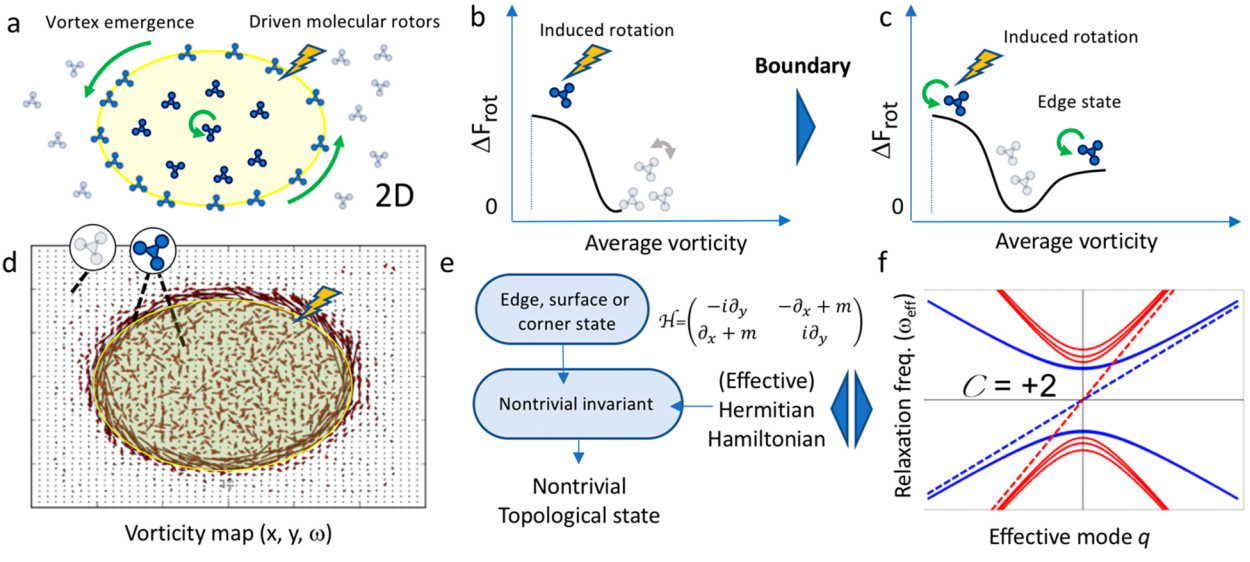

Figure 4. Topology of (nonequilibrium) thermodynamic states. (a) Vortex states emerge when molecular rotors or microspinners are driven due to coupling between rotational and translational degrees of freedom, and (b and c) may constitute the basis for future explorations of nonequilibrium topological thermodynamic edge states, see text. (d) Molecular dynamics and (e and f) mapping of the underlying hydrodynamic model onto an effective Dirac Hamiltonian, draw parallels to Chern insulators.[89] In the model, the "Dirac mass" is $m \sim \lambda^{-1}$, where λ is a hydrodynamic damping length arising from substrate friction. Adapted with permission from ref 89.

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the CI model,[16] provided a time-dependent perturbation is present. The corresponding topological edge states are also observed, consistent with a chiral hydrodynamic flow in both spin and vorticity (Figure 4).

The excitation of unidirectional molecular rotors for the engineering and control of topological chiral thermodynamic states can be technically challenging. An alternative is the mechanical transformation of linear momentum into angular momentum. This can be achieved by laminar vortex formation through chiral lattices,[91−93] similar to the acoustic effects[40] introduced in Figure 1c. Such experimental realizations pose challenges for incompressible and granular matter prone to overdamping, leading to jamming. Notwithstanding this, underdamped molecular systems at interfaces could overcome these disadvantages, thereby harnessing the notions of spin-CI models for paradigmatic topological function in soft matter at near-to-equilibrium conditions.

In summary, topological tools are now routinely employed in electronic, photonic, acoustic, and mechanical engineering. The virtues of topological quantum solids in chemistry are well-known,[94,95] yet the relevance of the topological classification of dynamic matter at finite temperatures is still the subject of intense theoretical research. Experimental surveys and simulations of physicochemical systems are expected to complement theoretical efforts, toward the demonstration of unconventional boundary states and properties of assemblies, reactions, and system chemistry. To this end, we proposed three levels of exploration: at the configurational topology level, where the study of dynamic topological defects and real-space reaction Hamiltonians is promising, yet the demonstration of nonlocal, nontrivial functionality is challenging; at the topological phononics level, where vibrational modes can be approximated by topological band theory under limited thermal fluctuations; and finally, topological thermodynamic states that are an intriguing theoretical and experimental concept, which can be phenomenologically studied by means of effective topological band models. Future work on the classification of topological thermodynamic states might benefit from exclusive topological tool development beyond condensed matter physics, aided by machine learning protocols which sample small deviations from equilibrium for example. Overall, the prospect of topological dynamic matter at finite temperatures is expected to stimulate the development of new directions and proof-of-principle studies in molecular and soft matter research.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.jpclett.0c03114.

SSH model description; polyacetylene models (Figure S1); band structure of trivial and nontrivial cases (Figure S2); contour plot (Figure S3) (PDF)

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Notes

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