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### Magnetism in curved geometries

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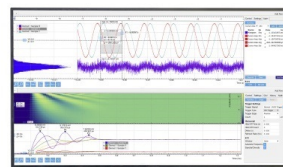
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## ABSTRACT

Curvature impacts physical properties across multiple length scales, ranging from the macroscopic scale, where the shape and size vary drastically with the curvature, to the nanoscale at interfaces and inhomogeneities in materials with structural, chemical, electronic, and magnetic short-range order. In quantum materials, where correlations, entanglement, and topology dominate, the curvature opens the path to novel characteristics and phenomena that have recently emerged and could have a dramatic impact on future fundamental and applied studies of materials. Particularly, magnetic systems hosting non-collinear and topological states and 3D magnetic nanostructures strongly benefit from treating curvature as a new design parameter to explore prospective applications in the magnetic field and stress sensing, micro-robotics, and information processing and storage. This Perspective gives an overview of recent progress in synthesis, theory, and characterization studies and discusses future directions, challenges, and application potential of the harnessing curvature for 3D nanomagnetism.

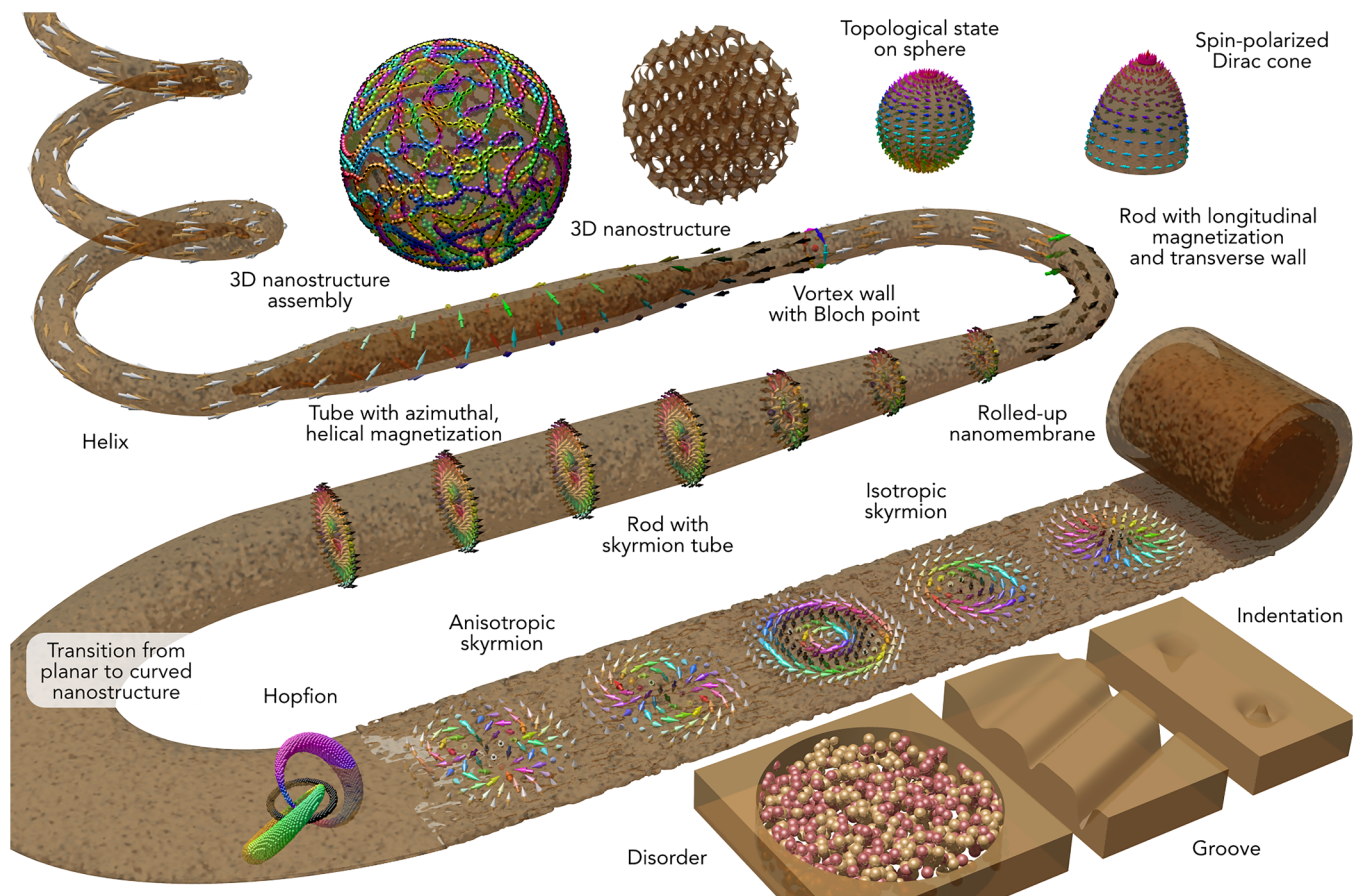
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## I. INTRODUCTION

Understanding the relationship between electronic and magnetic properties and structural and chemical quantities is one of the overarching themes in condensed matter and applied physics and key to the discovery of novel quantum materials. Correlated electron systems and magnetic materials are particularly interesting because microscopic characteristics of entangled and topological states are heavily determined by local atomic and nanoscale features. To date, research efforts have largely been focusing on synthesizing planar single-crystals, epitaxial films, and multilayer stacks with tailored functionalities originating from the nearly perfect long-range order and symmetry. The existence or absence of symmetry is essential to many phenomena emergent in topological insulators, ferroelectric, multiferroic, and magnetic materials whose physical properties are described by vector order parameters relying on, e.g., spin-orbit coupling.<sup>1–3</sup> In fact, current information processing and storage architectures as well as concepts for novel microelectronics, including the evolving field of spintronics,<sup>4</sup> rely on low-dimensional systems with well-defined symmetry and special types of spin-orbit coupling. However, structural and chemical inhomogeneities and disorder emerge even in the most perfect materials and at interfaces. A new way of describing those

imperfections is to assign them a curvature in real, reciprocal, or spin space (Fig. 1). A local curvature can be employed to design systems with spontaneous or inhomogeneous inversion symmetry breaking and to stabilize 3D magnetization vector fields or to tailor topology and magneto-transport properties in amorphous correlated electron systems.<sup>5–7</sup> Sculpting 3D curved nanostructures provides means to tailor the curvature on the nanoscale while simultaneously expanding 1D and 2D nanostructures into the third dimension<sup>8</sup> and is heavily used in microrobotics.<sup>9–11</sup>

Topological vector fields, such as vortices,<sup>12</sup> skyrmions<sup>13</sup> and topological knots,<sup>14–17</sup> possess a curvature in the vector order parameter space, e.g., spin space. Compared with uniformly polarized or topologically trivial configurations, topological vector fields span the Bloch sphere  $N$  times with  $N$  referring to the topological charge. The representation in terms of a Bloch sphere is convenient to describe electromagnetism in solids<sup>18–20</sup> and to link topological properties to electronic transport phenomena.<sup>21</sup> The latter has stimulated a multitude of theoretical and experimental studies of magnetic<sup>22–27</sup> and polar<sup>28,29</sup> skyrmions in a large variety of materials systems in view of both basic sciences and novel information storage and processing units, such as the racetrack memory.<sup>30–32</sup> Alternative concepts propose to use topological states as 3D curved



**FIG. 1.** Magnetism in curved geometries in real, reciprocal, and spin space. Magnetic properties and novel functionalities are governed by the curvature and short-range order alongside elements and composition. Local inversion symmetry breaking by the curvature, strain, and short-range order can promote the formation of 3D topological spin textures owing to an emergent local vector exchange interaction (DMI) with prospective applications to microelectronics while offering greater flexibility in materials synthesis. Geometrically confined structures, such as nanorods, nanotubes, and nanohelices, induce a curvature-driven DMI that discriminates between spin chirality and supports the nucleation of chiral and topological states with unprecedented stability upon current excitation. 3D nanostructures are synthesized by self-assembly of nanoparticles, nanoprinting, or etching enable microrobotics in gaseous and liquid phases, fundamental studies on 3D spin frustration, and 3D magnetic logic and storage systems.

magnonic waveguides<sup>33</sup> for spin wave-based spintronics,<sup>34</sup> for neuromorphic<sup>35–38</sup> and probabilistic<sup>39</sup> computing, or for topological magnonics,<sup>40,41</sup> taking advantage of their quasi-particle character. The vast majority of magnetic topological vector fields has been stabilized in systems with inversion symmetry breaking, provided either by virtue of their crystal structure<sup>42,43</sup> or through the presence of planar interfaces,<sup>44–46</sup> causing an asymmetric vector spin exchange known as the Dzyaloshinskii–Moriya interaction (DMI).<sup>42,43</sup> However, those concepts and governing mechanisms are universally applicable to ferroelectric,<sup>28,29</sup> multiferroic,<sup>47</sup> and 2D van der Waals materials,<sup>48–51</sup> as well as to amorphous materials<sup>52–54</sup> with local inversion symmetry breaking. In fact, systems with a locally varying DMI<sup>55,56</sup> or a spontaneous symmetry breaking with respect to spin chirality have been proposed for stabilizing twisted and anisotropic magnetic solitons,<sup>16,57</sup> including topological spin knots referred to as hopfions.<sup>14–17,58</sup> The inherent

dilemma of mutually exclusive small topological states and high magnetic ordering temperature, essential to spintronics applications, may be addressed using targeted synthesis of magnetically ordered alloys.<sup>59</sup> The prerequisite non-planar arrangement of atoms of the same element can be interpreted as a curved interface within the solid-state material, opening a completely new direction of exploring curvature as a new design parameter.

A complementary route to break inversion symmetry without impairing intrinsic properties relies on engineering curved nanostructures and tailoring magnetic exchange interactions.<sup>60</sup> Curvature has been employed to design tubular architectures with virtually unlimited magnetic domain wall velocity and unidirectional spin wave propagation owing to curvature-driven magneto-chirality.<sup>61</sup> Since curvature-driven inversion symmetry breaking is conceptually analogous to an emergent DMI, topological states can be created and manipulated solely by curvature<sup>62</sup> without the need

for intrinsic inversion symmetry breaking. To this extent, we envision that curvature will be used as a scientific design principle in the form of rough, curved structural, chemical, and magnetic interfaces, gradients, and inhomogeneities/disorder in solid-state materials. This includes, in particular, artificial magneto-electric materials,<sup>63</sup> local DMI to stabilize anisotropic topological states, room-temperature skyrmions spanning a few nanometers, and spin waves emanating from and along non-collinear spin textures, such as chiral domain walls<sup>64,65</sup> and 3D topological states,<sup>33</sup> enabling configurable 3D magnonic crystals. The advantage of designing and implementing curved vector fields over structurally predefined curvature opens a new way to tune on-demand the spin wave dispersion, i.e., band structure, through twisting and deforming or altering the topology of the magnetization configuration.

Similar to the recent success of expanding low-dimensional magnetism into 3D nanomagnetism,<sup>66–68</sup> implementing curvature as a design concept into future magnetic materials requires an integrated approach of advanced modeling, synthesis, and characterization to validate the properties and behavior of curved magnetic structures. Recent developments of analytical and numerical frameworks have allowed for quantifying curvature-induced magneto-chirality,<sup>61,69</sup> curvature-driven formation of topological states,<sup>62,70</sup> and vector spin exchange on the atomic scale.<sup>71</sup> Advances in electrochemical deposition<sup>72</sup> and 3D nanoprinting<sup>73–76</sup> enabled the synthesis of tubular, helical, and more complex nanostructures with ever-growing quality of magnetic and structural properties. Magnetic properties of planar films have been tailored by interface engineering taking advantage of improved growth capabilities and *ab initio* guided synthesis.<sup>3</sup> Magnetic microscopy, tomography, and scattering<sup>77–81</sup> at coherent x-ray light sources and aberration-corrected transmission electron microscopy centers have become essential to characterize chemical and structural inhomogeneities within the magnetic material and near interfaces/surfaces, and to visualize 3D magnetization vector fields. Great progress has been made in pushing limits of optical and scanning probe microscopies relying on, e.g., the Kerr effect, superconducting quantum interference device magnetometry,<sup>82</sup> and nitrogen-vacancy magnetometry.<sup>83</sup>

Given the enormous scientific opportunities and challenges with adding curvature as a critical parameter to magnetic materials, this Perspective provides an overview of recent progress in synthesis, theory, and experimental studies and discusses potential future directions of harnessing curvature for 3D nanomagnetism. In particular, we summarize the current state of 3D nanostructures, curvature effects, and their relation to topological magnetic states in Sec. II. Current and future technological advances in numerical modeling, synthesis, and characterization, enabling these scientific breakthroughs, are discussed in Sec. III. Sections IV and V give a scientific and technological perspective of the harnessing curvature for basic sciences and prospective applications of 3D nanomagnetism.

## II. STATE-OF-THE-ART OF CURVATURE-INDUCED EFFECTS

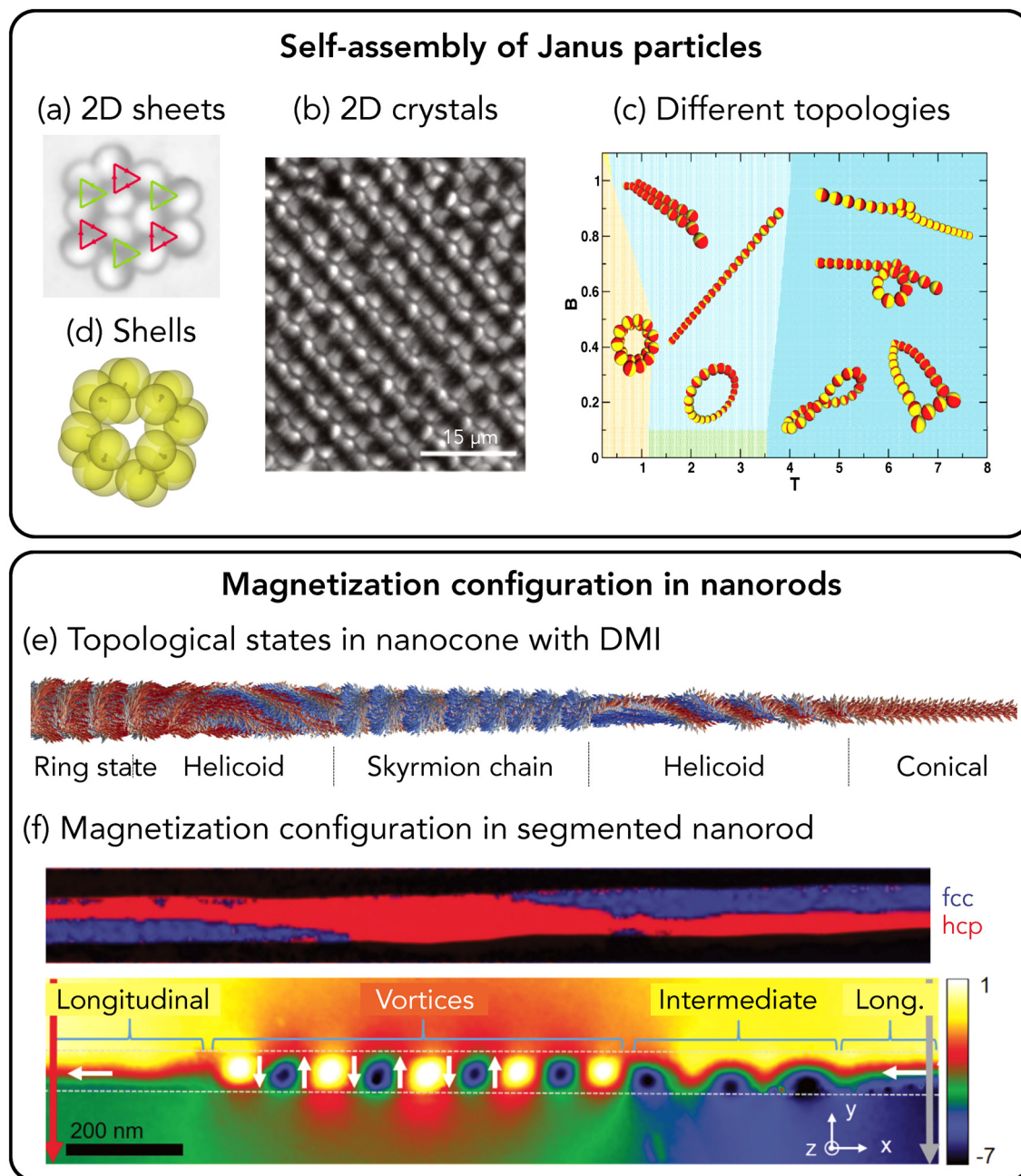
Curved geometries are characterized by the spatial distribution of the local inverse radius, i.e., curvature, which can span a wide range from  $1/\mu\text{m}$  down to  $10/\text{nm}$ . Generally, the upper and lower boundaries are governed by extrinsic properties, including the

shape and size of 3D nanostructures and structural deformation, and intrinsic properties, such as interfaces, heterogeneity, and disorder, respectively (Fig. 1). The unique feature of the curvature is its inherent local inversion symmetry breaking which, depending on its origin, leads to a constant or gradually/randomly changing modification to magnetic properties.<sup>60,84,85</sup> The former refers to the special case of a constant curvature and magnetization orientation with respect to the curvature; the latter to the general case of a varying microscopic or nanoscopic curvature. Note that this applies to real, reciprocal, and spin space; curved spin geometries in reciprocal and spin space affect mainly spin excitations and electronic transport due to different spin-orbit coupling phenomena. The effect of a locally varying curvature in the form of structural, chemical, electronic, and magnetic inhomogeneities and disorder scales with its ratio of magnitude to spatial variation. A sufficiently large ratio can affect magnetic properties and, for instance, stabilize topological spin textures on the corresponding length scale; otherwise, curvature-induced modifications to magnetic interactions will mostly compensate each other. On the other hand, engineering curved nanostructures allow for tailoring magnetic exchange interactions without impairing intrinsic properties. This approach is fundamentally different from traditionally tuning the shape and size to modify magnetic dipole energies of nanostructures.

### A. 0D and 1D nanostructures

The most prominent properties of nanostructures are the shape and size that alter or even completely suppress magnetism when approaching tens of nanometers. These modifications stem from an increased surface-to-volume ratio that boosts unfavorable magnetic dipole contributions, triggering a high sensitivity to short-range order and location/orientation of the magnetization of adjacent nanoparticles. The latter can be employed to design complex 3D nanostructure assemblies of core-shell and solid magnetic nanoparticles possessing a centered magnetic moment, as well as Janus particles with an off-center magnetic moment in the form of a well-defined in-plane or perpendicular<sup>86–88</sup> magnetization, vortices,<sup>89–92</sup> or topological states.<sup>93</sup> Magnetic short-range and even long-range orders manifest in clusters with spin frustration [Fig. 2(a)],<sup>94</sup> 2D heterostructured colloidal crystals [Fig. 2(b)],<sup>95</sup> and straight tubular chains with variable diameters.<sup>96</sup> Theoretical studies revealed novel assemblies beyond straight chains<sup>97</sup> and flakes, such as meandering chains,<sup>98</sup> rings with different sizes, shapes and topology [Fig. 2(c)],<sup>99–101</sup> and shells [Fig. 2(d)].<sup>102</sup>

Physically expanding a spherical nanoparticle along one axis results in cylindrical nanorods with a uniaxial structural and magnetic symmetry. These structures typically stabilize a longitudinal magnetization similar to planar nanowires lacking a magneto-crystalline anisotropy; all other magnetic properties, such as domain wall nucleation and motion, magnetization reversal, and spin wave propagation, are fundamentally different due to constant local curvature (circular cross section). Early theoretical works on domain wall nucleation and propagation in nanorods provided quantitative proof for a suppressed Walker breakdown<sup>103</sup> for transverse walls.<sup>104</sup> The unprecedented large domain wall velocities have only recently been contested by synthetic antiferromagnets and angular moment-compensated ferrimagnets<sup>105,106</sup> with inversion



**FIG. 2.** Magnetization configurations in 0D and 1D nanostructures. (a)–(d) Self-assembly of magnetic Janus particles into (a) and (b) 2D arrays with specific symmetry (static), (c) straight chains, closed loops, and helices (static, length, temperature, and field dependent), and (d) 3D shells (static, electric charge-stabilized). (a) Reproduced with permission from Baraban *et al.*, *Phys. Rev. E* **77**, 031407 (2008). Copyright 2008 American Physical Society. (b) Reproduced with permission from Tsyrenova *et al.*, *Langmuir* **35**, 6106–6111 (2019). Copyright 2019 American Chemical Society. (c) Reproduced with permission from Hernández-Rojas and Calvo, *Phys. Rev. E* **97**, 022601 (2018). Copyright 2018 American Physical Society. (d) Reproduced with permission from D. Morphew and D. Chakrabarti, *Nanoscale* **10**, 13875 (2018). Copyright 2018 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (e) 3D non-collinear spin textures forming in FeGe nanorods with DMI revealing dependence on spatial confinement (diameter). Reproduced with permission from Charilaou and Löffler, *Phys. Rev. B* **95**, 024409 (2017). Copyright 2017 American Physical Society. (f) Magnetization in Co-rich CoNi nanorods with face-centered cubic (fcc) and hexagonal close packed (hcp) crystal structures visualized with electron holography. Reproduced with permission from Andersen *et al.*, *ACS Nano* **14**, 1399 (2020). Copyright 2020 American Chemical Society. (a), (b), and (f) and (c)–(e) are experimental and numerical data, respectively.

symmetry breaking. The combination of uniaxial symmetry and shape of the nanorods causes, depending on the diameter,<sup>107</sup> either deterministic nucleation of transverse domain walls or Bloch points at the center of vortex walls<sup>108–110</sup> (Fig. 1). More complex non-collinear spin textures emerge in nanorods with an intrinsic inversion symmetry breaking and resulting in Dzyaloshinskii–Moriya interaction (DMI),<sup>42,43</sup> which reveal a periodic switching between skyrmionic and helical spins for matching and non-matching rod diameter, respectively [Fig. 2(e)].<sup>111</sup>

In recent years, tremendous progress has been made in synthesis and visualizing the magnetization configuration,<sup>112</sup> including magnetization reversal process and correlation with local structural and chemical properties. Adopting x-ray photon emission electron microscopy<sup>113</sup> to conduct transmission experiments and analyzing both direct and shadow XMCD contrast enabled the visualization of helical spins in curved nanomembranes<sup>114</sup> and nanorods<sup>115</sup> and studying 3D printed nanohelices.<sup>116</sup> The correlation between local structural and chemical properties and the magnetization configuration has been addressed with electron holography in nanorods in terms of imperfections,<sup>117–119</sup> such as grains and surface roughness, and engineered chemical/structural segmentation [Fig. 2(f)].<sup>120–122</sup> The latter approach allowed for transforming a simple longitudinal magnetization prevailing in elongated nanorods into helical, vortex, or transverse configurations, which are strong contenders for novel spin torque nanooscillators. Recently, experimental studies of the current-driven domain wall motion in nanorods corroborated the theoretically predicted high velocities.<sup>123</sup>

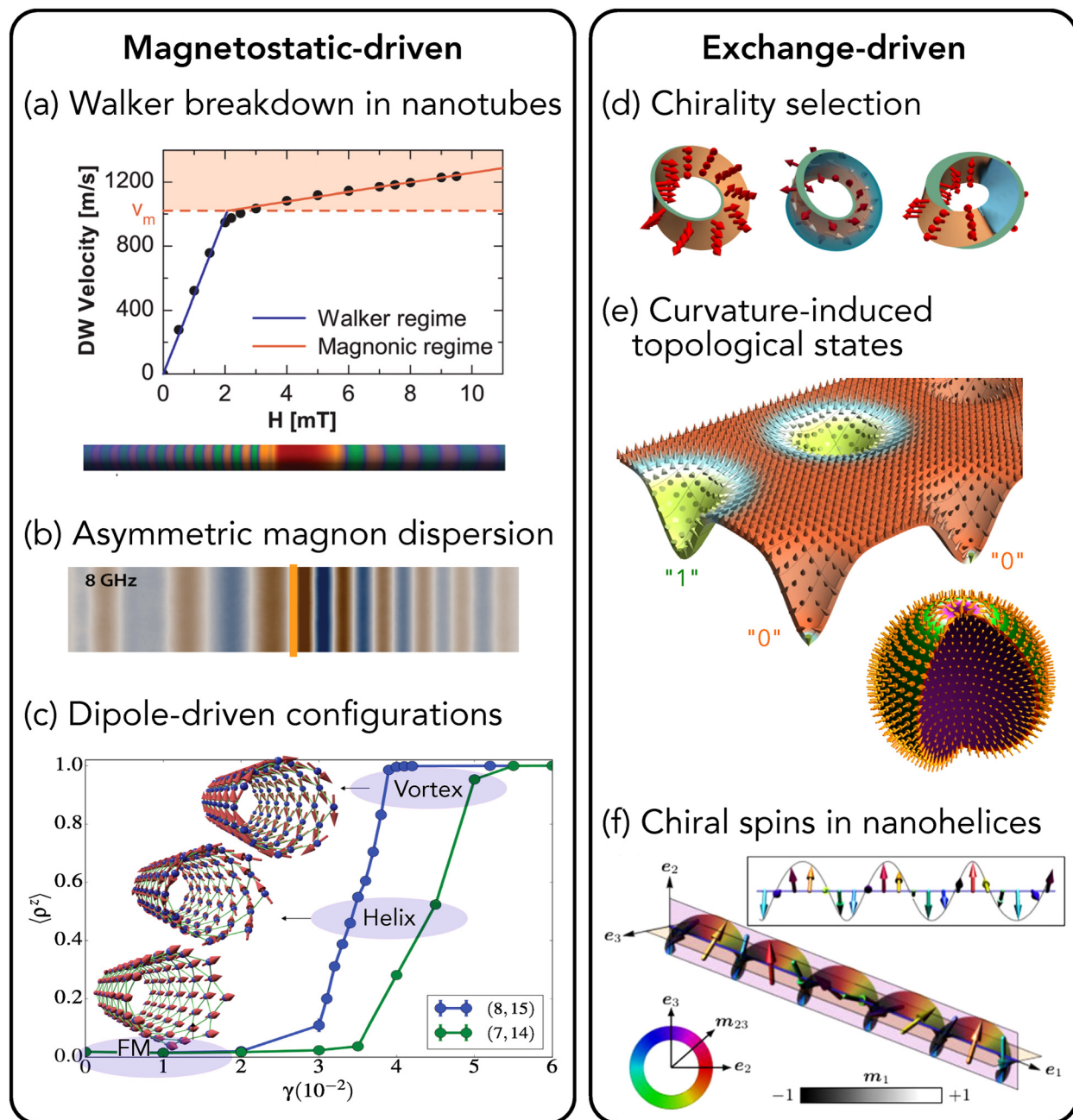
## B. 2D curved geometries and curvature effects

Nanoparticles (0D) and nanorods (1D) without a magnetic core resemble shell (spherical shell) or ring (nanotube) structures with distinct magnetic properties are governed by topology and curvature. Hollow tubular architectures with longitudinal magnetization and vortex domain walls lack, in contrast to nanorods, a Bloch point in the center (Fig. 1). These nanostructures promise virtually unlimited magnetic domain wall velocity [Fig. 3(a)],<sup>124</sup> unidirectional spin wave propagation [Fig. 3(b)],<sup>125–128</sup> and vortex chirality-dependent standing spin wave spectra,<sup>127,129</sup> owing to curvature-driven magneto-chirality.<sup>61,69</sup> The latter refers to the spin chirality selection in nanotubes due to lifted degeneracy between moving vortex walls with opposite circulation. The spin transfer torque tilts the magnetization within the domain wall inward (outward), depending on its circulation, and enhances (impairs) the stability of the vortex wall by reducing (increasing) its magnetic stray field.<sup>125</sup> In addition to these dynamic modifications, tube diameter or strength of magnetic dipole interactions can be varied to switch between longitudinal, helical and vortex configurations [Fig. 3(c)].<sup>130</sup> Engineering systems with unidirectional spin wave propagation are appealing for energy efficient magnonics<sup>34</sup> and creating unidirectional magnetoacoustic waves.<sup>131</sup> The former has just recently been demonstrated in planar architectures by resonantly exciting non-collinear spin textures, such as vortices,<sup>65</sup> Bloch points,<sup>64</sup> and domain walls,<sup>132</sup> in ferromagnetic and synthetic antiferromagnets. Particularly interesting is the spin wave propagation along curved domain walls<sup>64</sup> and skyrmion tubes<sup>33</sup> to design 3D reconfigurable magnonic waveguides.

Mathematically, curved geometries can be treated as planar systems following a coordinate transformation. The generalized theory of curvilinear micromagnetism<sup>60</sup> illustrates how local and non-local interactions emerge from the curvature, including a magnetic exchange interaction similar to DMI (Fig. 4),<sup>66,84,85</sup> an easy-surface anisotropy,<sup>133</sup> and, for rough interfaces and surfaces and heterogeneous materials, i.e., local curvature, a spatial distribution of easy-axis, easy-cone, or easy-plane anisotropy. The vector spin exchange originates from the local inversion symmetry breaking and causes a local preference for spin chirality if the magnetization and normal vector of the curved surface are not aligned (Fig. 4). For instance, spheres and tubes with radial magnetization do not show a preference. The curvature-driven DMI puts magneto-chirality<sup>69</sup> in a broader context and explains the emergence of chiral and topological spin textures in curved surfaces with cylindrical symmetry,<sup>134</sup> cones,<sup>84</sup> twisted bands,<sup>135</sup> Möbius bands [Fig. 3(d)],<sup>136</sup> tori,<sup>137</sup> bent nanotubes<sup>138</sup> and rods,<sup>139,140</sup> nanohelices,<sup>63,141,142</sup> shells,<sup>143–146</sup> and indentations [Fig. 3(e)].<sup>62,70</sup> Antiferromagnetic nanohelices support the formation of coherent magnon condensates in the momentum space.<sup>142</sup> Geometrically tailoring the curvature of nanohelices allows for stabilizing topologically distinct chiral spin textures, such as cycloidal and helicoidal configurations, as well as collinear single-domain and multi-domain states [Fig. 3(f)].<sup>63,141</sup> These states can be transformed into each other by stretching or squeezing the nanohelix, offering a new approach to design magneto-electric materials without external magnetic fields.<sup>63</sup> Similarly, the vortex ground state in ring-shaped nanowires transitions upon deformation into the trivial onion state.<sup>147</sup> These transformations represent an unwinding of chiral, topological spin textures into trivial states, triggered by the curvature-induced DMI.

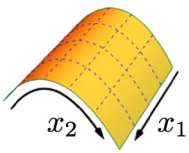
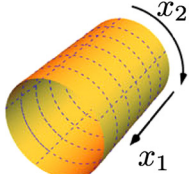
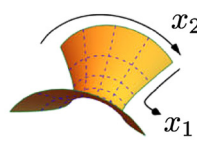
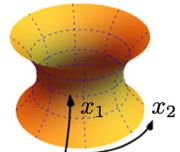
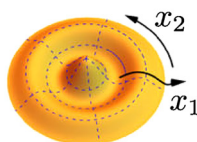
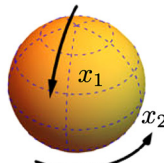
Nanoscale indentations enable the stabilization and manipulation of topological spin textures, such as skyrmions and skyrmioniums also known as target skyrmions [Fig. 3(e)],<sup>62,70</sup> in magnetic materials with otherwise absent inversion symmetry breaking. Relying on exchange instead of magneto-static energies, this mechanism is fundamentally distinct from using thickness gradients to nucleate vortex lattices in percolated non-planar films.<sup>148</sup> The shape, size, and topology of the magnetic state can be tailored by adjusting the local curvature. In a broader sense, these theoretical studies infer that local inversion symmetry breaking, due to structural and chemical inhomogeneity, and rough, curved interfaces causes an inhomogeneous, local DMI in real materials. The challenge is to engineer these curved interfaces to promote the formation of topological spin textures instead of randomly canted spins occurring in frustrated spin systems.

To date, experimental studies of curvature effects are still rare. Magnetic switching in tubular architectures with radial magnetization and nanotubes with longitudinal magnetization were visualized with magnetic force microscopy<sup>149</sup> and superconducting quantum interference device microscopy.<sup>82</sup> These tabletop tools, providing spatial resolution on the sub-100 nm scale, represent a significant advancement compared with earlier works using cantilever magnetometry.<sup>150,151</sup> Azimuthal soft-magnetization configurations,<sup>152</sup> inaccessible in planar geometry, have proven essential to giant magneto-impedance field sensors with unprecedented sensitivity.<sup>153</sup> A first glimpse of the potential of curvature for spintronics was recently given by disentangling spin and charge resistance in



**FIG. 3.** Numerically predicted curvature effects in 2D curved geometries. (a) Ultra-fast domain wall velocities in nanotubes due to delayed Walker breakdown associated with spin chirality selection. Reproduced with permission from Yan *et al.*, *Appl. Phys. Lett.* **99**, 122505 (2011). Copyright 2011 AIP Publishing LLC. (b) Asymmetric magnon dispersion in nanotubes originating from spin chirality selection similar to interfacial DMI in planar systems. Reproduced with permission from Otálora *et al.*, *Phys. Rev. Lett.* **117**, 227203 (2016). Copyright 2016 American Physical Society. (c) Transformation of magnetic states in nanotubes with magnetic dipole coupling strength. From Salinas *et al.*, *Sci. Rep.* **8**, 10275 (2018). Copyright 2018 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (d) Spin chirality selection in Möbius bands governed by perpendicular magnetic anisotropy. From Pylypovskyi *et al.*, *Phys. Rev. Lett.* **114**, 197204 (2015). Copyright 2015 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (e) Formation of skyrmions in nanoindentations and spherical surfaces due to emergent DMI associated with local inversion symmetry breaking. Reproduced with permission from Kravchuk *et al.*, *Phys. Rev. Lett.* **120**, 067201 (2018). Copyright 2018 American Physical Society and *Phys. Rev. B* **94**, 144402 (2016). Copyright 2016 American Physical Society. (f) Helicoidal spin textures in nanohelices reversibly transforming into homogeneous and periodical states upon stretching/compression. From Volkov *et al.*, *Sci. Rep.* **8**, 866 (2018). Copyright 2018 Author(s), licensed under a Creative Commons Attribution (CC BY) license.



Curved geometry	Anisotropy		Local interactions			Non-local interactions
	Type	$\mathbf{e}_a$	$w_A^x$	$w_{D1}^x$	$w_{D2}^x$	
	easy-axis	$\hat{\mathbf{n}}, \mathbf{e}_2$	✓	—	✓	✓
	hard-axis	$\hat{\mathbf{n}}$	✓	—	—	—
	easy-axis	$\mathbf{e}_1$	✓	—	—	—
	easy or hard-axis	$\mathbf{e}_a$	✓	—	✓	✓
	easy-axis	$\hat{\mathbf{n}}$	✓	—	—	✓
	hard-axis	$\mathbf{n}$	✓	—	—	—
	easy-axis	$\mathbf{e}_1$	✓	—	—	—
	easy-axis	$\mathbf{e}_2$	✓	—	—	—
	easy or hard-axis	$\mathbf{e}_a$	✓	✓	✓	—
	easy-axis	$\hat{\mathbf{n}}, \mathbf{e}_1$	✓	✓	✓	—
	easy or hard-axis	$\mathbf{e}_a$	✓	✓	✓	✓
	easy-axis	$\hat{\mathbf{n}}$	✓	—	—	✓
	easy or hard-axis	$\mathbf{e}_a$	✓	✓	✓	✓

**FIG. 4.** Theory of curvilinear micromagnetism. Non-local magnetic interactions emerge from a curvature-driven DMI in systems where the magnetization is not aligned along the normal vector of the curved surface. From Sheka *et al.*, *Commun. Phys.* **3**, 128 (2020). Copyright 2020 Author(s), licensed under a Creative Commons Attribution (CC BY) license.

aluminum nanowires deposited above a groove.<sup>154</sup> The experiment showed a higher efficiency compared with planar structures, which is essential to low-power spin current electronics and 3D microelectronics architectures with curved interconnects.

### C. Topological states

Topological magnetic states are 3D inherently curved magnetization vector fields (Fig. 1) that behave like quasi-particles upon magnetic and electric field excitation.<sup>155</sup> The magnetic properties and electronic transport phenomena<sup>21</sup> are linked via solid-state electromagnetism,<sup>18–20</sup> which sets them apart from chiral domain walls and vortices<sup>12</sup> and makes them both fundamentally intriguing and relevant to low-power spin-based microelectronics. Since the theoretical prediction of skyrmions<sup>13</sup> and their experimental observation in single-crystals<sup>156,157</sup> and thin films,<sup>158</sup> magnetic skyrmions have been extensively studied in view of current manipulation,<sup>159</sup> current creation,<sup>160,161</sup> and electric detection of individual skyrmions via the topological Hall effect<sup>162,163</sup> associated with the perpendicular deflection of skyrmions<sup>164</sup> or, more recently, the Nernst effect.<sup>165,166</sup> These investigations had mainly been driven by engineering planar interfaces<sup>3</sup> to tailor spin-orbit coupling, essential to DMI<sup>42,43</sup> (formation), topological Hall effect (detection), and spin-orbit torque (manipulation). Synthesizing ultra-thin multilayer stacks with tailored interfacial DMI<sup>71</sup> enabled the stabilization of room-temperature skyrmions in ferromagnets<sup>167</sup> and ferrimagnets.<sup>106,168,169</sup> The latter benefit from significantly enhanced current-driven velocities near angular moment compensation. Using complex oxide materials to grow epitaxial interface heterostructures with broken inversion symmetry and a large gradient of the electrostatic potential promoted the formation of skyrmions at low temperature<sup>170,171</sup> whose size can be controlled by the ferroelectric polarization.<sup>172</sup>

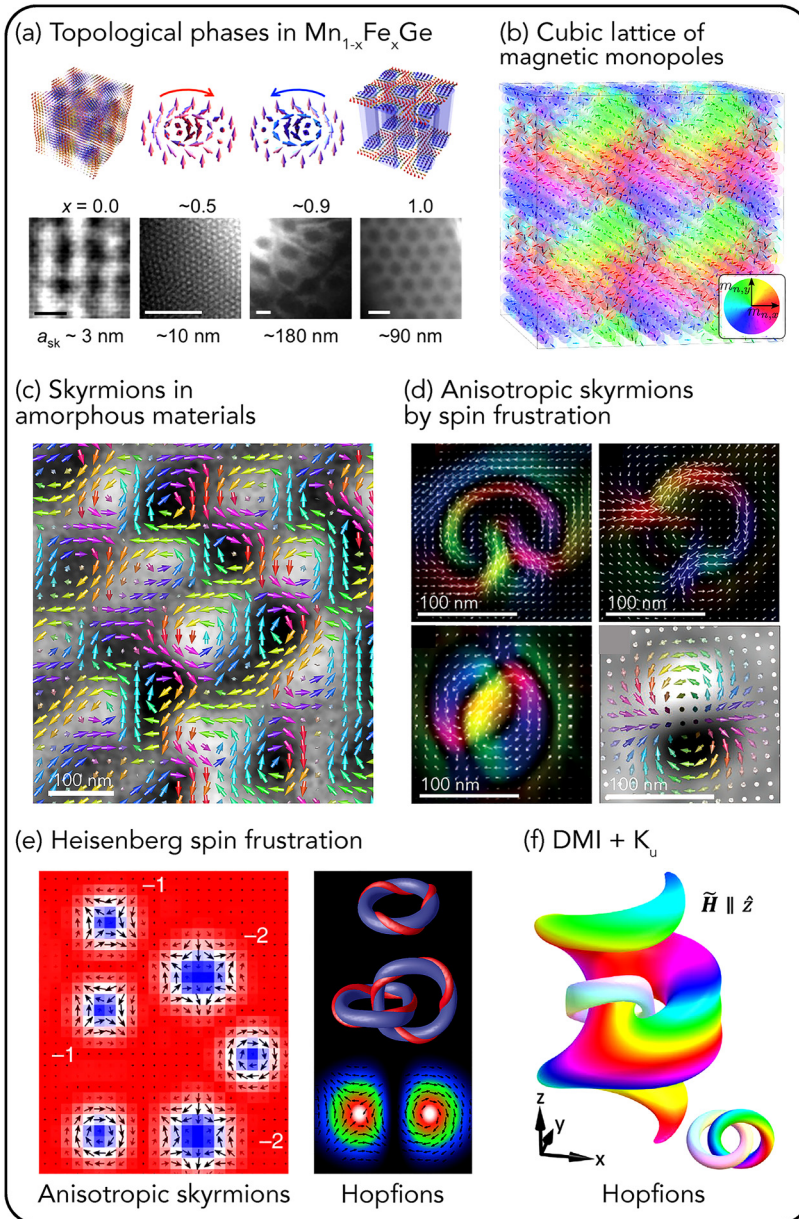
The smallest room-temperature skyrmionic spin structures (<20 nm) were stabilized by pseudo-random substitution of Si atoms with Co excess atoms in polycrystalline B20 Co-Si materials.<sup>59</sup> Disorder also exists in multicomponent B20 single-crystals stabilizing topological phases [Fig. 5(a)],<sup>173</sup> such as  $\text{Fe}_{1-y}(\text{Co}, \text{Mn})_y(\text{Ge}, \text{Si})$ ,<sup>59,173–176</sup> which crystallize as a chiral lattice with an inherent chemical disorder that becomes chiral to the atomic building blocks.<sup>177</sup> A recent theoretical work<sup>178</sup> showed the necessity of spin frustration to explain the experimentally observed transition between different topological phases in B20 structures [Fig. 5(b)],<sup>173,179</sup> including magnetic monopoles on the order of 1 nm,<sup>179–181</sup> and rebuked the commonly accepted requirement of large DMI to stabilize small topological states. Atomistic simulations<sup>182</sup> and experimental studies<sup>183,184</sup> of amorphous ferrimagnets confirmed further the persistence of DMI in structurally and chemically disordered materials. In fact, chemical and structural disorder can cause bulk DMI<sup>52,184</sup> and stabilize topological states in amorphous compounds [Fig. 5(c)].<sup>54</sup> This is attributed to an increased Anderson localization<sup>185,186</sup> and the suppression of electron transfer between transition metal atoms that enlarge local density of states and spin-orbit coupling,<sup>187</sup> local DMI, magneto-resistance, and Hall effects.<sup>188</sup> However, observing topological knots in polycrystalline soft-magnetic bulk materials lacking inversion symmetry breaking demonstrated a certain degree of randomness in the occurrence of topological objects similar to magnetic vortices in

extended soft-magnetic films.<sup>58</sup> The coordination number of the amorphous structure can be tuned by the deposition temperature from a high-coordination-phase at low temperatures<sup>189</sup> to a lower-coordination-phase at room temperature<sup>190</sup> with a short-range order resembling that of B20 structures. In this context, disorder refers to locally varying DMI due to atomic short-range order and not to randomly distributed pinning sites, which have theoretically been investigated in view of current-driven skyrmion dynamics.<sup>191–195</sup>

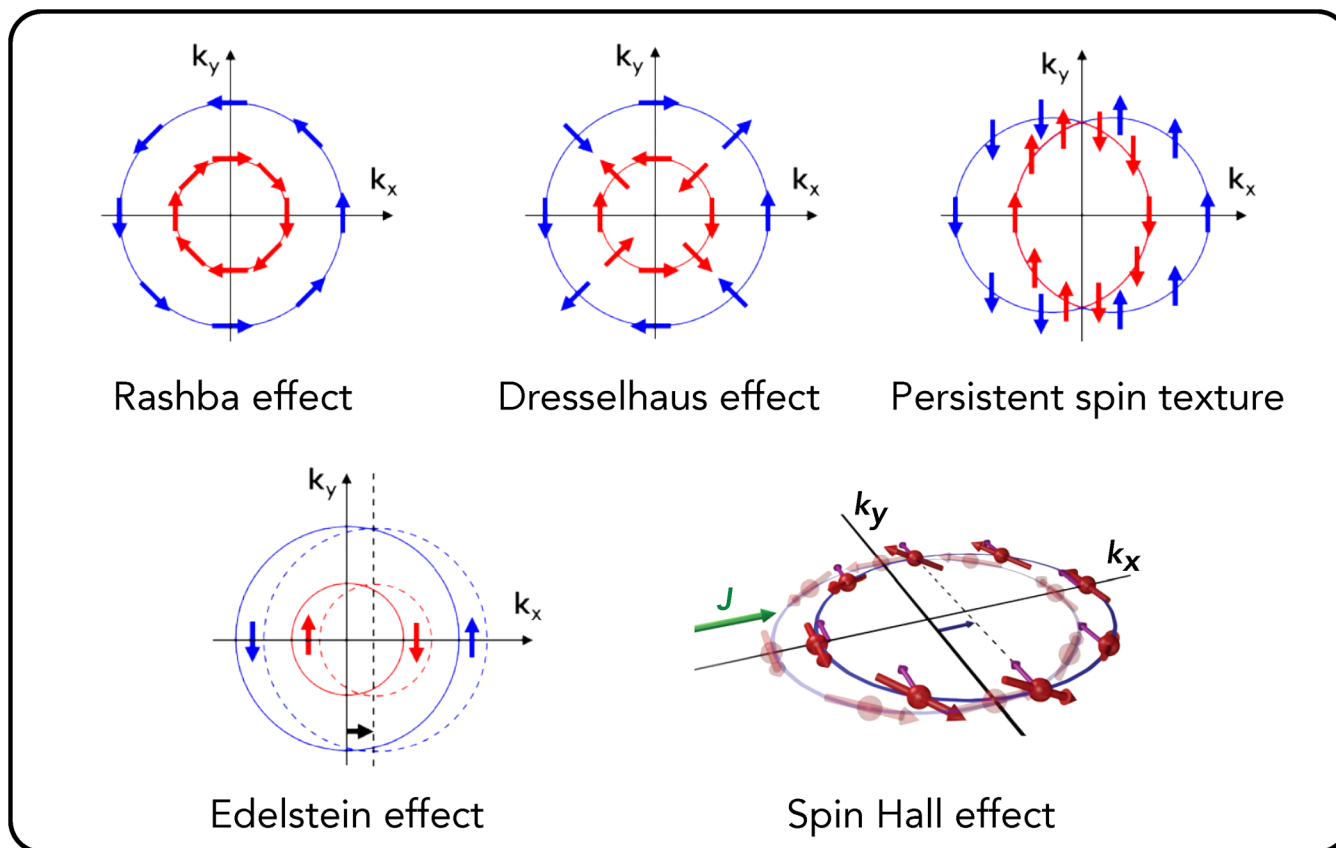
Systems with a locally varying DMI<sup>55,56</sup> or a spontaneous symmetry breaking with respect to spin chirality, triggered by spin frustration, have been proposed for stabilizing twisted and anisotropic magnetic solitons,<sup>56,57</sup> including topological spin knots referred to as hopfions<sup>16,17,196</sup> [Fig. 5(e)], and, in part, experimentally been observed [Fig. 5(d)].<sup>54,58,197,198</sup> The lowest-order hopfion can be pictured as a spin torus which is twisted along its circumference continuously transforming between vortex and antivortex. The increased complexity of hopfions generally hinders a deterministic formation; a recent numerical study proposed to combine spatial confinement with DMI and perpendicular magnetic anisotropy to stabilize hopfion-like spin textures [Fig. 5(f)].<sup>196</sup> These higher-order, anisotropic topological states possess a vanishing gyro-vector and intrinsically compensate the perpendicular deflection of quasi-particles due to Magnus force promising a straight trajectory at increased velocities [Figs. 11(b) and 11(c)]. Examples range from biskyrmions (bound pair of skyrmions with opposite chirality)<sup>199–201</sup> and bilayer skyrmions<sup>202</sup> to antiskyrmions,<sup>203,204</sup> skyrmioniums (biaxial skyrmions)<sup>205–207</sup> and antiskyrmioniums<sup>208</sup> to skyrmion bags,<sup>209,210</sup> and hopfions,<sup>211,212</sup> as well as antiferromagnetic topological states.<sup>213–217</sup> Moreover, the Magnus force can be suppressed by nanoscale modifications to structural and magnetic properties in the form of tracks and pinning sites,<sup>23,218</sup> or switching to tubular systems with a corresponding helical skyrmion trajectory.<sup>219</sup> Considering topological states in 2D and disordered materials further benefits novel concepts for manipulating magnetic exchange and topological states via curvature [Fig. 3(e)],<sup>62,70</sup> voltage,<sup>220–225</sup> strain,<sup>226–228</sup> or pressure,<sup>229–231</sup> which are less effective or even destructive in (poly-)crystalline metallic systems. These alternate routes provide a convenient way to twist and deform or even alter the topology of 3D curved magnetization vector fields needed to design configurable 3D magnonic crystals<sup>33</sup> or tunable topological magnonics.<sup>40,41</sup>

### D. Curved spin geometries in reciprocal space

In addition to magnetic exchange manifesting collinear, non-collinear, and topological magnetism, spin-orbit coupling enables an efficient charge-to-spin conversion and current-induced spin-orbit torques, mediated by non-trivial spin textures in reciprocal space. The latter originate from inversion symmetry-breaking Dresselhaus<sup>232</sup> and Rashba<sup>233</sup> fields, which impose a spin chirality on the electronic bands (Fig. 6) and generate a non-equilibrium spin polarization. The conversion between charge and spin current relies on the (inverse) Edelstein or (inverse) spin Hall effect and has experimentally been observed at, e.g., non-magnetic metal interfaces<sup>234,235</sup> and insulating oxide interfaces,<sup>236,237</sup> and in ferroelectric materials<sup>238</sup> and 2D van der Waals heterostructures.<sup>239</sup>



**FIG. 5.** Topological states stabilized by inversion symmetry breaking and spin frustration. (a) Topological phase transitions between trigonal and cubic skyrmion phases in B20  $\text{Mn}_{1-x}\text{Fe}_x\text{Ge}$  with  $x$  visualized with Lorentz microscopy (electron intensity shown). From Kanazawa *et al.*, *New J. Phys.* **18**, 045006 (2016). Copyright 2016 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (b) *Ab initio* calculations of MnGe based on Heisenberg spin frustration without DMI revealing cubic lattice of skyrmions and magnetic monopoles. Reproduced with permission from Mendive-Tapia *et al.*, *Phys. Rev. B* **103**, 024410 (2021). Copyright 2021 American Physical Society. (c) Coexistence of helical spins and skyrmions in amorphous Fe–Ge films visualized with Lorentz microscopy (electron phase and magnetization depicted). Reproduced with permission from Streubel *et al.*, *Adv. Mater.* **33**, 2004830 (2021). Copyright 2021 John Wiley and Sons. (d) Anisotropic skyrmions in (left)  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ , transforming into each other via field-driven motion of Bloch lines and (right) amorphous Fe–Ge retrieved from Lorentz microscopy. Reproduced with permission from Yu *et al.*, *Adv. Mater.* **29**, 1603958 (2017). Copyright 2017 John Wiley and Sons and from Streubel *et al.*, *Adv. Mater.* **33**, 2004830 (2021). Copyright 2021 John Wiley and Sons. (e) Formation of anisotropic skyrmions and topological knots (hopfions, preimages, and cross section shown) by Heisenberg spin frustration retrieved from micromagnetic simulations. From Zhang *et al.*, *Nat. Commun.* **8**, 1717 (2017). Copyright 2017 Author(s), licensed under a Creative Commons Attribution (CC BY) license and Sutcliffe, *Phys. Rev. Lett.* **118**, 247203 (2017). Copyright 2020 American Physical Society. (f) Metastable hopfion relaxed in system with DMI and uniaxial symmetry by micromagnetic simulations (preimages shown). Reproduced with permission from Balasubramanian *et al.*, *Phys. Rev. Lett.* **125**, 057201 (2020). Copyright 2020 American Physical Society. (a), (c), and (d) and (b), (e), and (f) are experimental and numerical data, respectively.



**FIG. 6.** Spin-orbit coupling phenomena with corresponding spin orientation of the two spin-split electronic sub-bands for systems without inversion symmetry. The Edelstein effect causes a spin accumulation due to shifted Fermi surfaces with an external electric field. An electric current  $J$  displaces the Fermi surface along its flow direction, thereby tilting their spins up (down) for  $k_y > 0$  ( $k_y < 0$ ) and creating a spin current in the  $y$ -direction (spin Hall effect). L. L. Tao and E. Y. Tsybal, Nat. Commun. **9**, 2763 (2018). Copyright 2018 Author(s), licensed under a Creative Commons Attribution (CC BY) license and Sinova *et al.*, Rev. Mod. Phys. **87**, 1213 (2015). Copyright 2015 American Physical Society.<sup>392</sup>

Topological materials possess a gapless surface state protected by time-reversal symmetry,<sup>240</sup> whose band dispersion features a Dirac cone decorated with electron spins pointing tangential to the surface.<sup>241</sup> This spin texture is dramatically changed upon magnetic doping due to local time-reversal symmetry breaking that opens a gap at the Dirac point and causes a magnetically induced hedgehog-like spin configuration (Fig. 1).<sup>242</sup> The latter allows for generating spin currents<sup>243</sup> and producing spin-transfer torques on adjacent ferromagnets.<sup>242</sup> Chiral crystals, which lack inversion, mirror, or other rotation-inversion symmetries, stabilize topologically non-trivial spin textures whose spin components parallel to the electron momenta appear around highly symmetric  $k$ -points.<sup>244</sup> Both existence and inverted topology in right- and left-handed crystals were recently observed in chiral tellurium crystals,<sup>241</sup> promising pure spin current generation. In ferroelectric materials, the spin texture is coupled to and can be controlled by the ferroelectric polarization providing a promising platform to explore the coupling between spin, orbital, valley, and lattice degrees of freedom in solids.<sup>245</sup>

One particular benefit of Rashba spin-orbit coupling is its control by a gate voltage across an interface supporting a 2D electron gas in the form of a spin field-effect transistor. Its practical realization is challenging since non-collinear spin textures possess a reduced spin diffusion length owing to an enhanced magnetic impurity and defect scattering of electrons changing their momentum and randomizing the spin.<sup>246</sup> This effect can be circumvented by engineering structures where the magnitudes of Rashba and Dresselhaus spin-orbit coupling are equal, resulting in a unidirectional spin-orbit field and a momentum-independent spin configuration, known as the persistent spin texture (Fig. 6). Under these conditions, the electron motion is accompanied by spin precession around the unidirectional spin-orbit field, leading to a spatially periodic mode referred to as a persistent spin helix.<sup>247</sup> The latter is robust against spin-independent disorder and offers an infinite spin lifetime. It has experimentally been demonstrated in a 2D electron gas semiconductor quantum-well structure by tuning quantum-well width and doping,<sup>247,248</sup> and theoretically been predicted in bulk oxide materials with a non-symmorphic space group

symmetry.<sup>249</sup> Examining the role of local curvature, structural, and chemical disorder in view of real-space inversion symmetry breaking and their potential to enhance charge-to-spin conversion and increase critical temperatures is critical particularly to the emergent field of topological amorphous materials.<sup>5</sup>

### III. TECHNOLOGICAL ADVANCES

Scientific advances and future directions are heavily intertwined with technological advances in numerical modeling, synthesis and characterization. Their development throughout the last decade has diversified both the original research perspective and scientific community.

#### A. Numerical modeling

Accurate modeling of magnetic systems and magnetic interactions in solid-state materials is more important than ever before to accelerate materials discovery, predict magnetic phase transitions, static, and dynamic properties, and refute or corroborate analytical and experimental data. This is mainly because of an increased complexity compared to, e.g., planar ferromagnetic Permalloy ( $\text{Ni}_{80}\text{Fe}_{20}$ ) films, and driven by the use of multiple elements, including transition metal, metalloids, heavy-element, and rare-earth materials, curved nanostructures, disorder, and link to spin-orbit coupling and magneto-transport phenomena. Affordable parallel computing with multi-core central processing units (CPU) and graphics processing units (GPU), as well as inexpensive memory have helped establish numerical simulations as a mainstream technique to address curvature phenomena.

Numerous public-domain finite element/difference method software packages are available to model magnetic materials with DMI, including legacy OOMMF,<sup>250</sup> MuMax3,<sup>251</sup> and Fidimag.<sup>252</sup> Atomistic solvers, such as Vampire,<sup>253</sup> Spirit,<sup>254</sup> and Fidimag,<sup>252</sup> enable a more accurate modeling of singular magnetic spin textures, e.g., Bloch points/lines and skyrmions, antiferro- and ferrimagnetism, helimagnets, and frustrated systems, as well as 3D curved nanostructures, structural and chemical disorder, and temperature effects. Micromagnetic simulations of 3D curved geometries with arbitrary shape can be carried out with Nmag,<sup>255</sup> which is a powerful framework in combination with the HLib library.<sup>256</sup> Future developments will accommodate computational intense calculations of elastic properties and magnetostriction, and time-dependent deformation and motion of realistic multifunctional materials. One leap in this direction has been done by Boris Computational Spintronics,<sup>257</sup> a multi-physics software with incorporated heat flow solver, electronic transport solver, temperature-dependent material parameters, and mechanical stress-strain solver. While these micromagnetic platforms offer insight into magnetic states, magnetization reversal processes, size effects, and current- and field-driven spin excitations, they do rely on physical parameters such as saturation magnetization, magnetic exchange interactions, magnetic anisotropy, etc., typically retrieved from experiments or *ab initio* calculations.

In the wake of interface and curvature engineering, density functional theory is essential to determining the dependence of interface and curvature effects, including DMI and spin-orbit torque, on used elements, and structural and chemical order. Three of the most popular *ab initio* frameworks are FLEUR,<sup>258</sup> VASP,<sup>259</sup> and Quantum ESPRESSO,<sup>260</sup> which provide means to model band structures and

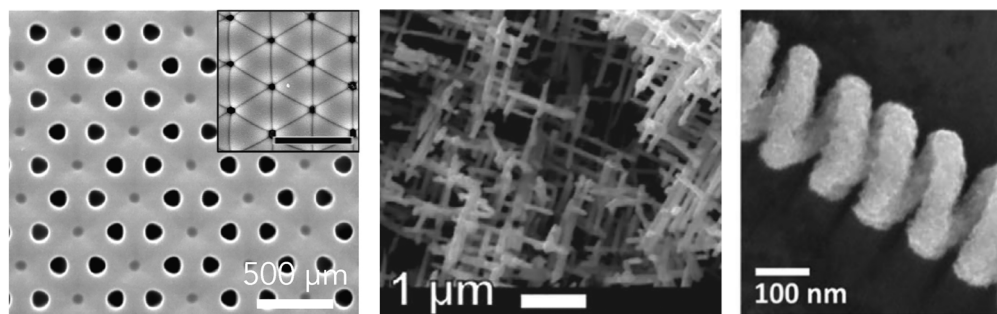
quantify atomic DMI values. The numerical results of exchange-coupled systems strongly depend on the atomic coordinates, which are typically approximated according to their crystalline structures. However, this presumption is invalid for inhomogeneous and disordered materials. Arguably, the actual coordinates of each individual atom are virtually impossible to determine; the systems can however be approximated according to their short-range order that can be quantified with molecular dynamics simulations using, e.g., LAMMPS.<sup>261</sup> The latter simulates dynamic processes of assembly, nucleation and diffusion during synthesis or upon external stimulation on the atomic scale. This hierarchical approach of modeling will become more important to future studies of real materials with imperfections, disorder and highly inhomogeneous regions, including amorphous materials and interfaces.

#### B. Synthesis

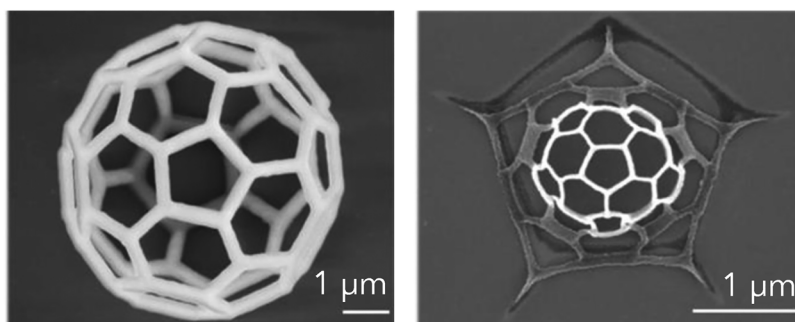
Engineering interfaces has been a focus of recent research on nanomagnetic materials, primarily due to the possibility to harness the spin-orbit coupling induced by symmetry breaking effects at such interfaces.<sup>2,3</sup> These efforts have been guided by *ab initio* calculations to identify the best pairing of heavy-element material or oxide and magnetic element in view of largest DMI values<sup>71</sup> to stabilize chiral spin textures and topological states. Magneto-transport properties, such as the spin Hall effect and spin-orbit torque, essential to current manipulation of chiral spin textures have typically been phenomenologically optimized and correlated to *ab initio* calculations. In conjunction with exploring different classes of materials, including atomic monolayers, epitaxial, polycrystalline and amorphous films as well as 2D materials, this approach has flourished owing to employing both intrinsic and extrinsic (interface) properties, which offers new functionalities and greater flexibility in materials synthesis. The next decade will show to which extent structural and chemical inhomogeneity, disorder, and curved interfaces can be harnessed to tailor magnetic exchange interactions and manipulate topological states in solid-state materials.

The synthesis of 3D nanostructures utilizing electrochemical deposition [Fig. 7(a)],<sup>72,262,263</sup> two-photon lithography [Fig. 7(b)],<sup>75,264–267</sup> and focused electron beam-induced deposition [Fig. 7(c)]<sup>73,74</sup> has seen tremendous progress particularly with respect to controlling shape, roughness, morphology, homogeneity and purity. Electrochemical deposition using porous alumina or gyroid polymer<sup>268</sup> templates enabled the synthesis of nanorods, nanotubes nanohelices, multi-segmented specimens,<sup>115,117–122,269</sup> and 3D networks<sup>270?–272</sup> with variable diameter (<50 nm), length ( $\sim 1\mu\text{m}$ ), and metallic materials [Fig. 7(a)]. The default trigonal symmetry of the porous template was circumvented by focused ion beam guided anodization.<sup>270</sup> Focused electron beam-induced deposition has taken the lead in synthesizing 3D nanostructures with virtually any shape and curvature, including nanowires,<sup>116,274,275</sup> networks [Fig. 7(c)],<sup>276–278</sup> and topological structures.<sup>279</sup> Relying on the dissociation of adsorbed metal-cabonyl precursor molecules by the electron beam, the printed metallic nanostructures typically incorporate carbon or oxygen impurities of  $\geq 10\%$ , which can be reduced using reactive gases during synthesis or post-growth. In-depth studies of process parameters, such as growth rate,

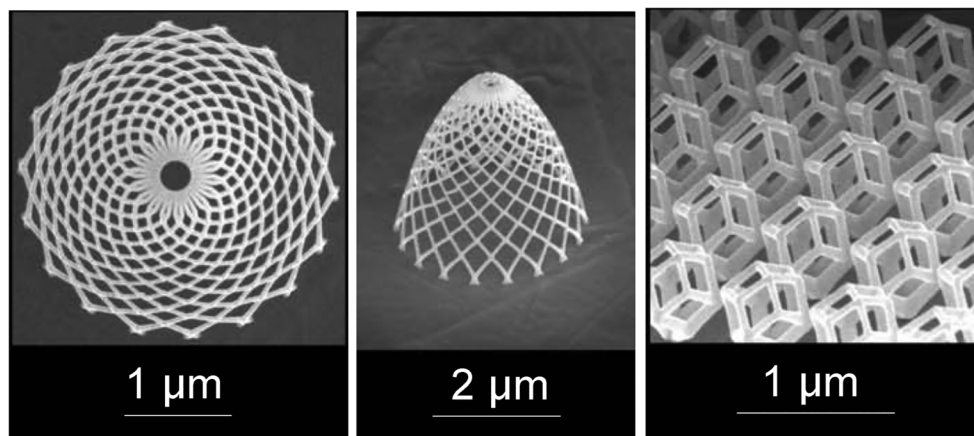
## (a) Electrochemical deposition



## (b) Two-photon lithography



## (c) Electron beam-induced deposition



**FIG. 7.** Synthesis of 3D nanostructures using bottom-up techniques. (a) Electrochemical deposition into porous alumina templates for fabrication of magnetic nanorod arrays, 3D magnetic networks, and nanohelices. Reproduced with permission from Chen *et al.*, *Langmuir* **27**, 800 (2011). Copyright 2011 American Chemical Society; from Wagner *et al.*, *Adv. Electron. Mater.* **7**, 2001069 (2021). Copyright 2021 Author(s), licensed under a Creative Commons Attribution (CC BY) license; and reproduced with permission from Sattayasamitsathit *et al.*, *Nanoscale* **6**, 9415 (2014). Copyright 2014 Royal Society of Chemistry. (b) Polymer templates obtained by two-photon lithography (left) and additional etching and pyrolysis (right) revealing significant shrinkage. Reproduced with permission from Seniutinas *et al.*, *Microelectron. Eng.* **191**, 25 (2018). Copyright 2018 Elsevier. (c) Nanoprinting of 3D nanostructures utilizing electron beam-induced deposition through dissociation of metal-carbonyl precursor molecules. From Janbaz *et al.*, *Sci. Adv.* **3**, eaao1595 (2017). Copyright 2017 Author(s), licensed under a Creative Commons Attribution (CC BY) license.

precursor depletion/diffusion and heat load,<sup>277,280</sup> and computer-aided nanofabrication,<sup>278,281</sup> including Monte Carlo simulations of reaction–diffusion processes, have been essential to advancing 3D nanoprinting and investigating curvature and topology effects in 3D nanostructures.

Alternate techniques for 3D nanostructuring are printing polymeric 3D nanotemplates with two-photon lithography and subsequent metal deposition,<sup>264</sup> implosion fabrication harnessing shrinkage and dehydration of hydrogel scaffolds,<sup>282</sup> and self-assembly of nanoparticles on curved liquid–liquid interfaces to structure liquids<sup>283–285</sup> that can be endowed with a remanent magnetization (Fig. 10).<sup>76,286,287</sup> Nanoindentations with engineered curvature can be carved out via dry etching with ion irradiation prior to non-epitaxial film deposition. Post-growth nanoscale modifications to magnetic exchange, anisotropy and saturation magnetization may be performed with low-current ion irradiation.<sup>218</sup> Another versatile technique with respect to tailored magnetic properties is strain engineering rolled-up nanotech<sup>288–290</sup> that facilitates internal strain gradients to manufacture tubular magnetic geometries with variable diameter/curvature and thickness.<sup>152,291,292</sup> Subsequently, strain engineering has been generalized to synthesize shape-morphing micromachines (Fig. 13),<sup>293–296</sup> reconfigurable actuators,<sup>297,298</sup> and shape memory polymers<sup>299</sup> with magnetic functionality.

### C. Characterization

Whether 3D nanostructures or topological magnetic states, the challenge with characterizing 3D magnetization vector fields is the complexity and ambiguity of many characterization techniques due to the lack of knowledge about all three magnetization components and their spatial distribution at a sufficient spatial resolution. Joint studies harnessing multimodal techniques and subsequent detection of remaining components have provided means to identify stable magnetic states. The most advanced tools with respect to resolution and sensitivity are x-ray and electron techniques, complemented by tabletop instruments, such as scanning probe and optical microscopy, magneto-transport, electron spin resonance spectroscopy, and magnetic neutron scattering revealing internal spin structures of nanoparticles.<sup>300,301</sup> Choosing state-of-the-art instrumentation is typically a compromise between high sensitivity, high spatial resolution, temporal resolution, and accessibility. Additional constraints are element specificity, interaction between probe and magnetization, and environment, e.g., applying current/voltage, strain/pressure and magnetic fields or changing temperature and gas/solutions, to create, manipulate and detect magnetic states.

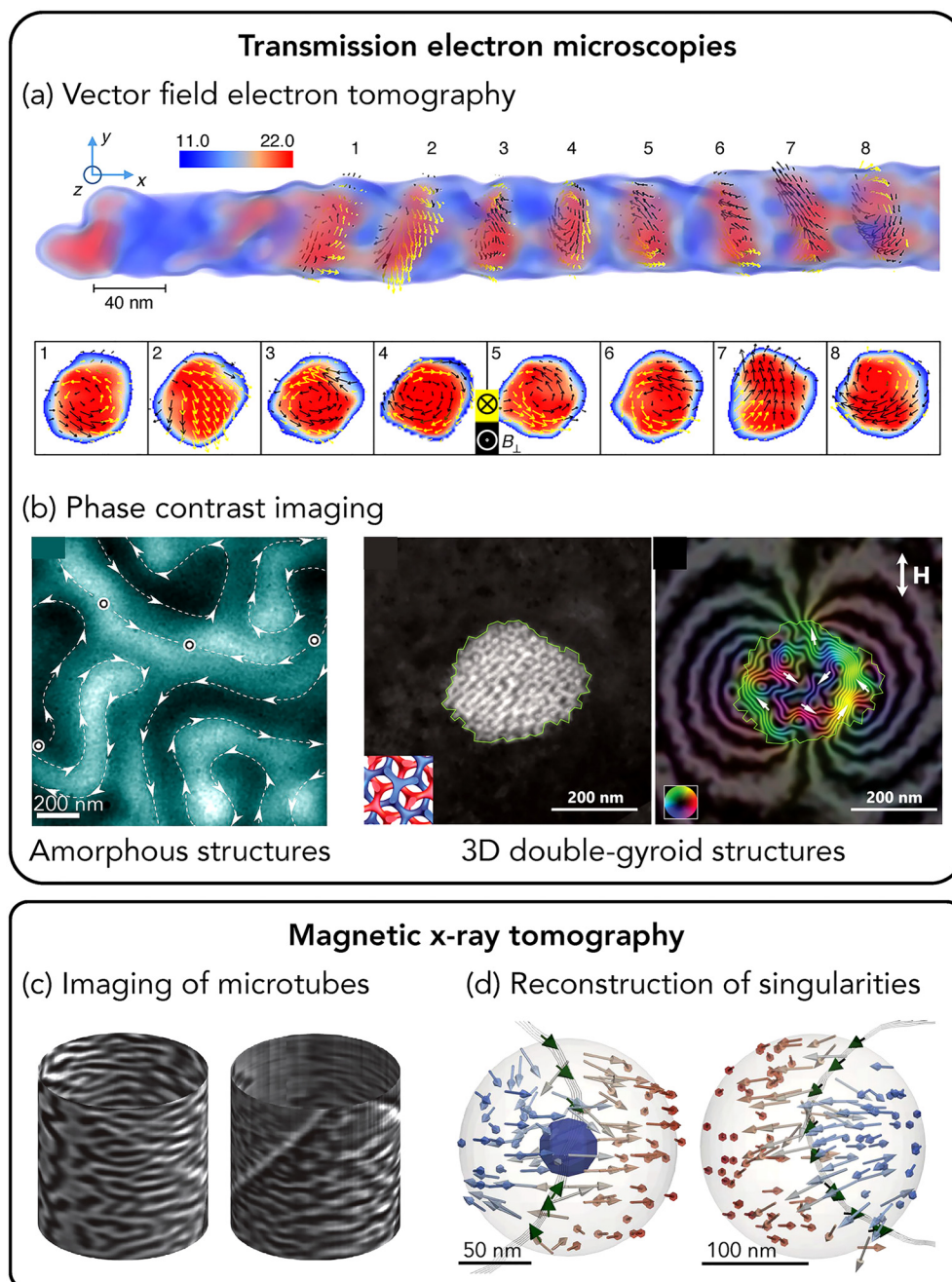
#### 1. Advanced electron and x-ray characterization

Electron microscopy<sup>80,81</sup> combines subatomic spatial resolution and beam coherence. One prime example harnessing both quantities is atomic scalar tomography to examine atomic order, internal defects, and strain of nanoparticles in vacuum<sup>302–305</sup> and liquid cells.<sup>306–308</sup> An adequate technique to visualize the magnetization on the atomic scale would tremendously benefit the study of antiferromagnets, disordered materials and topological states spanning only a few atoms. Future demonstrations might be accomplished by recording the diffraction pattern using 4D scanning transmission electron

microscopy similar to current approaches for strain mapping.<sup>304</sup> For now, 3D magnetization vector fields can be reconstructed on the nanoscale using vector field tomography based on off-axis or in-line electron holography [Fig. 8(a)].<sup>122,309,310</sup> Electron holography allows for studying the interaction of electromagnetic waves with 3D nanostructures<sup>117–119,311</sup> or thin films<sup>54,157,183,197,205,312–315</sup> on the nanoscale [Fig. 8(b)]. In-line holography, also known as Fresnel mode Lorentz microscopy, retrieves the electron phase from a focal plane series using transport-of-intensity equation<sup>316</sup> or Gerchberg–Saxton algorithm<sup>317</sup> without the need for a biprism and reference beam. The latter can also be avoided by leveraging differential phase contrast.<sup>318–320</sup> Magnetic (vector) and electrostatic/structural (scalar) contributions to the electron phase can be separated by subtracting the phase of the magnetically saturated state or of the flipped sample. A third option unique to the Gerchberg–Saxton algorithm takes into account different length scales and phase amplitudes<sup>54,183</sup> as well as the slow convergence of low-frequency components of non-electrostatic features during the iterative phase retrieval.<sup>321</sup> The simultaneous detection of both in-plane components of the magnetic induction (two-dimensional gradient of the electron phase) sets electron microscopy apart from x-ray techniques and is essential to time-resolved studies of the vast majority of magnetic systems.

X-ray spectromicroscopies,<sup>79</sup> harnessing x-ray magnetic circular dichroism or x-ray magnetic linear dichroism as element-specific absorption contrast mechanism, have been a workhorse for quantifying orbital and spin moments<sup>322,323</sup> and visualizing magnetization configurations and spin excitations on the tens of nanometer scale. Time-resolved measurements concerned thermal spin fluctuations, and current- and magnetic field-driven nucleation and manipulation of chiral domain walls, topological states and magnons. Orbital moments offer insight into the local electron orbital alignment. The limitation to one magnetization component can be overcome, for stable magnetic states, by tilting the sample to access another component or performing vector field tomography. Within the last few years, magnetic x-ray tomography has matured from prototypical demonstration with soft x rays [Fig. 8(c)]<sup>292</sup> to full-scale soft and hard x-ray tomography [Fig. 8(d)]<sup>58,324–326</sup> to stroboscopic tomographic imaging of driven magnetization dynamics.<sup>327</sup> This incredible progress has been possible by algorithm and hardware development at numerous synchrotron facilities. Alternatively, resonant x-ray scattering<sup>328</sup> can be employed to determine periodicity, spin chirality, and depth profile of periodic structures, such as skyrmion lattices in magnetic<sup>329,330</sup> and ferroelectric<sup>331</sup> materials. The interference pattern under the magnetic diffraction peak created by coherent x rays was used to reconstruct aperiodic magnetization vector fields on the nanoscale harnessing ptychography<sup>77,78</sup> and to study thermal spin fluctuations near, e.g., topological phase transitions on the nanosecond time scale with x-ray photon correlation spectroscopy at free electron laser facilities.<sup>332</sup>

Phase contrast imaging, such as x ray and electron ptychography, holography, and tomography techniques, is based on wave propagation and offers superior sensitivity and contrast compared with conventional microscopy. At synchrotron facilities, a coherent x-ray beam is currently generated by a pinhole smaller than 10  $\mu\text{m}$  that clips more than 90% of the beam. Free electron lasers and



**FIG. 8.** Advanced electron and x-ray characterization of 3D curved geometries. (a) Magnetization in Co/Cu multilayered nanorods obtained from holographic vector field electron tomography. From Wolf *et al.*, *Commun. Phys.* **2**, 87 (2019). Copyright 2019 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (b) Electron phase contrast imaging of (left) chiral ferrimagnetism in amorphous structures and (right) spin frustration in 3D printed nanostructures visualized with Lorentz microscopy and electron holography, respectively. Reproduced with permission from Streubel *et al.*, *Adv. Mater.* **30**, 1800199 (2018). Copyright 2018 John Wiley and Sons and Llandro *et al.*, *Nano Lett.* **20**, 3642 (2020). Copyright 2020 American Chemical Society. (c) 3D imaging of radial magnetization in tubular Co/Pd microstructures using soft x rays. From Streubel *et al.*, *Nat. Commun.* **6**, 7612 (2015). Copyright 2015 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (d) The 3D reconstruction of magnetic singularities in striped domain patterns in  $\text{Ni}_{80}\text{Fe}_{20}/\text{NdCo}_5/\text{Ni}_{80}\text{Fe}_{20}$  films. From Hierro-Rodríguez *et al.*, *Nat. Commun.* **11**, 6382 (2020). Copyright 2020 Author(s), licensed under a Creative Commons Attribution (CC BY) license.



field-emission aberration-corrected transmission electron microscopes already provide a coherent x ray and electron beam, respectively. Ongoing developments of faster, more sensitive detectors, and better optics and sources, e.g., monochromatic, brilliant, smaller, and coherent beams, offered by aberration-corrected transmission electron microscopes and next-generation diffraction-limited light sources, will significantly lower data acquisition time and improve illumination conditions and accessibility to a limited number of high-end instruments. Answering scientific questions concerning the relation between magnetization, structural and chemical order, topology and electric response will strongly rely on advancing operandi and time-resolved capabilities ranging from millisecond (pump-free)<sup>54,333</sup> to picosecond (pump-probe)<sup>334,335</sup> time scales. The former is limited by the detector, the latter by the pulse width and pulse separation. Nanofabrication advances include developing platforms for current, voltage and piezoelectric strain manipulation, and correlating magneto-transport properties, such as the topological Hall effect, with the magnetization configuration of individual topological states. The latter will benefit from increasing the number of aberration-corrected transmission electron microscopes and x-ray beamline endstations with liquid helium cryostat holders and allow for studying topological states in topological insulators and magnetic systems with possible quantum fluctuations. Ambient experiments using, e.g., oxygen and hydrogen gas provide means to modify interface/surface chemistry (spin-orbit coupling) or exchange interaction by reversible hydrogen intercalation. Liquid cells based on amorphous silicon nitride nanomembranes or graphene can be used to control magnetism via chemical means or study self-assembly of nanoparticles.

## 2. Tabletop instrumentation

Magneto-optical Kerr effect magnetometry and microscopy,<sup>336</sup> offering sensitivity to normal, transverse, or longitudinal magnetization components of the outer 20 nm provided means to study magnetization reversal processes and current-driven manipulation of micrometer-sized topological states<sup>161</sup> and 3D nanostructures by analyzing reflections from different surface regions [Fig. 9(a)].<sup>291,337–339</sup> These measurements can be combined with magneto-transport experiments to retrieve magneto-resistance and (topological) Hall coefficient for non-collinear spin textures [Fig. 9(b)],<sup>181</sup> or with micro-Hall probes to determine the magnetic hysteresis loops for the entire 3D nanostructures [Fig. 9(c)].<sup>281,340</sup> Magnetic imaging on the tens of nanometer scale is commonly carried out with magnetic force microscopy<sup>341</sup> by probing the second derivative of the normal stray field. Recent advances in magnetic tip customization have enabled simultaneous measurements of multiple components,<sup>342</sup> a configurable tip magnetization to track normal and in-plane components,<sup>343</sup> nanotube-based monopole sensors,<sup>344</sup> and a significantly enhanced sensitivity.<sup>345</sup> These steps are essential to quantitative magnetic force microscopy and reconstructing the 3D magnetization vector field. The challenge with magnetic force microscopy is the magnetic dipole interaction between tip and sample that alters the states in soft-magnetic systems or drags magnetic domain walls in relatively hard-magnetic materials. This limitation has been addressed with non-invasive scanning probe microscopies. Superconducting quantum interference device microscopy<sup>82,346</sup> measures the magnetic induction on the

nanoscale, which was demonstrated with ferromagnetic nanotubes and nanocubes [Fig. 9(d)]. Nitrogen-vacancy scanning probe microscopy<sup>83,347</sup> emerged as a highly sensitive technique to image non-collinear spin textures at the nanoscale in 2D van der Waals materials<sup>348</sup> and antiferromagnets<sup>349</sup> and reconstruct the full 3D magnetization vector field.<sup>350</sup> Extended magnetic phases were classified with respect to topology and chirality of the 3D spin textures using electron spin resonance spectroscopy.<sup>231,351,352</sup> The latter also allows for quantifying magnetic exchange stiffness and damping. Leveraging the magnon dispersion in inversion symmetry-broken systems, Brillouin light scattering provided means to quantify DMI near the interface, including spin chirality inversion in ferrimagnets.<sup>184</sup>

## IV. SCIENTIFIC PERSPECTIVE

Growing expertise and capabilities in modeling, synthesis and characterization will enable researchers to explore rich sciences not only in magnetism and condensed matter physics, but also in close conjunction with engineering, biology, and chemistry where the magnetization is either of central importance or a mean to improve functionality. In 3D nanomagnetism, this is reflected in an increasing number of theoretical and experimental works that harness curvature in 3D nanostructures and real, disordered materials to manipulate topological states.

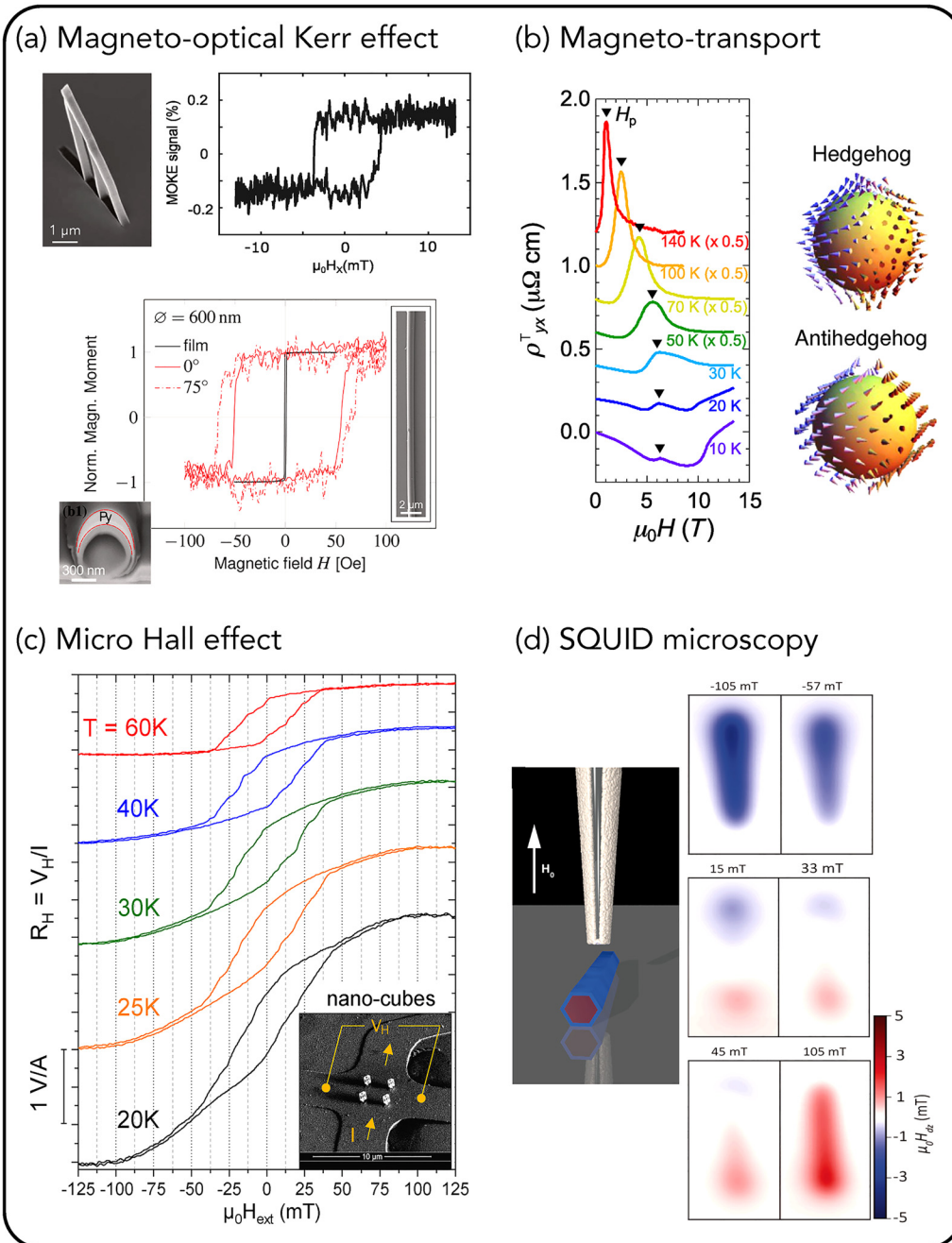
### A. 3D nanostructures

We anticipate three major research directions concerning assemblies of nanoparticles, spin frustration in 3D nanostructures, and magnetization vector fields and spin excitations in curved geometries.

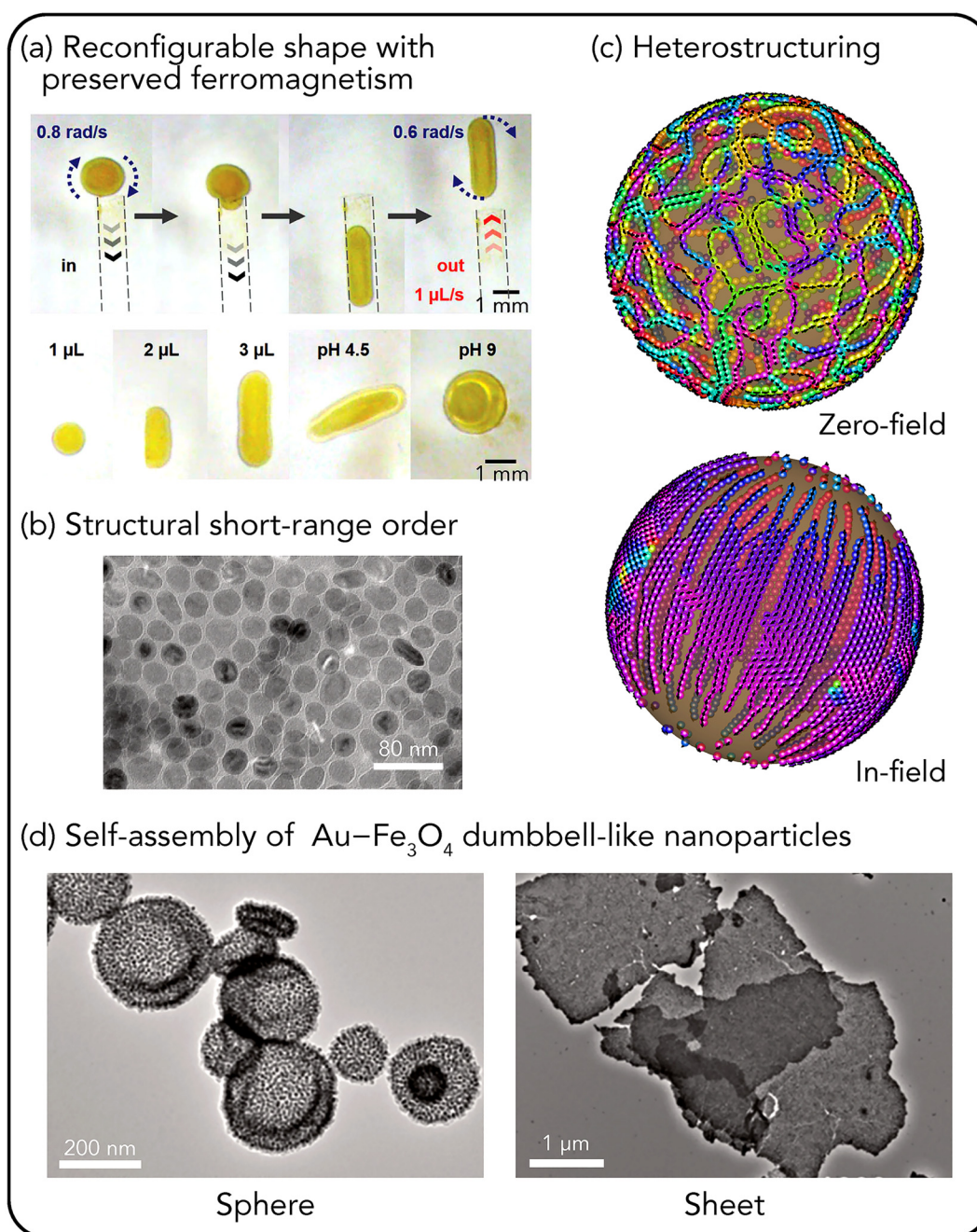
#### 1. Nanoparticle assembly

Experimental investigation of theoretically predicted nanoparticle assemblies with non-trivial geometries, such as shells, rings, helices, and nanopatterns with different sizes, shapes, and topology will face challenges with the inherent size and shape distribution of nanoparticles that cause disorder and deformation of the assembly. Numerical modeling will need to account for these experimental limitations to provide better insight into the self-assembly and its application potential. Disordered particle crystals and curved dipole-coupled systems can be used to explore disorder and curvature effects and to determine to which extent dipole systems resemble exchange-coupled materials.

While the vast majority of experimental and theoretical studies of nanoparticle assembly in solution<sup>353</sup> has relied on magnetic dipole interactions, there is ample opportunity to employ mechanical (gravity, surface tension) and chemical (pH, ligands) means. One prominent example is the assembly and jamming of nanoparticles at inversion symmetry broken liquid-liquid interfaces, which provide a reversible structural transformation between liquid and glassy states [Figs. 10(a) and 10(d)].<sup>76,354</sup> The glassy state can be pictured as a skeleton enclosing the liquid core with potentially high anisotropic, non-equilibrium shape. In combination with superparamagnetic nanoparticles, this approach enables a transition between paramagnetic ferrofluid and ferromagnetic liquid housing 2D ferromagnetism on the curved liquid interface.<sup>76,286,287,355</sup> With each



**FIG. 9.** Tabletop characterization tools for 3D nanomagnetism. (a) Surface-sensitive magneto-optical Kerr effect magnetometry of 3D conduit nanoprinted by focused electron beam-induced deposition of Permalloy and tubular Permalloy cap structure. Reproduced with permission from Sanz-Hernández *et al.*, ACS Nano **11**, 11066 (2017). Copyright 2017 American Chemical Society and Streubel *et al.*, Nano Lett. **12**, 3961 (2012). Copyright 2012 American Chemical Society. (b) Magneto-transport in B20 MnGe single-crystal showing temperature-dependent topological Hall effect due to emergence of topological states, including magnetic monopoles shown on the right. Reproduced with permission from Kanazawa *et al.*, Phys. Rev. Lett. **125**, 137202 (2020). Copyright 2020 American Physical Society. (c) Micro-Hall effect measurements on CoFe nanocube frames printed by focused electron beam-induced deposition. From Al Mamoori *et al.*, Materials **11**, 289 (2018). Copyright 2018 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (d) Superconducting quantum interference device microscopy visualizing the magnetization reversal process in ferrimagnetic nanotubes. Reproduced with permission from Vasyukov *et al.*, Nano Lett. **18**, 964 (2018). Copyright 2018 American Chemical Society. All techniques can be applied to 3D curved geometries.



**FIG. 10.** Self-assembly of nanoparticle building blocks into 3D hierarchical systems in liquid environment. (a) Interfacial jamming of superparamagnetic nanoparticles at liquid–liquid interfaces forming ferromagnetic liquid droplets with reconfigurable shape and preserved magnetization observed in hydrodynamics experiments. Reproduced with permission from Liu *et al.*, *Science* **365**, 264 (2019). Copyright 2019 American Association for the Advancement of Science. (b) Isotropic elastic and magnetic properties stemming from structural short-range order of jammed nanoparticles imaged with transmission electron microscopy in their dried state. Reproduced with permission from Wu *et al.*, *Proc. Natl. Acad. Sci. U.S.A.* **118**, e2017355118 (2021). Copyright 2021 National Academy of Sciences. (c) In-field and zero-field assembly of nanoparticles from a mixture of dispersed superparamagnetic and non-magnetic nanoparticles revealing distinct heterostructures with enhanced magnetic anisotropy and remanent magnetization. The depicted states are modeled with molecular dynamics and micromagnetic simulations and leave out non-magnetic nanoparticles. (d) Self-assembly of  $\text{Au-Fe}_3\text{O}_4$  dumbbell-like nanoparticles with packing parameter of (left) 0.63 and (right) 0.84 visualized with transmission electron microscopy. Reproduced with permission from Liu *et al.*, *Nano Lett.* **20**, 8773 (2020). Copyright 2020 American Chemical Society.

nanoparticle acting as a uniformly magnetized macrospin, thermal spin excitations, magnetic short-range, and long-range order depend on the structural short-range order of adjacent nanoparticles [Fig. 10(b)],<sup>286</sup> similar to macrospins in planar systems.<sup>356,357</sup> These systems have the potential to become a versatile platform to investigate spin liquid to spin glass transitions in planar and curved geometries which can be controlled by chemical properties, such as pH and ligands, stress and strain, electric and magnetic fields. The pH affects the screening of electrostatic charges and thus the separation between negatively charged nanoparticles jammed at the interface. Jamming mixtures of non-magnetic and superparamagnetic nanoparticles in the presence of an external magnetic field provides a route to adaptive reconfigurable 3D printing and heterostructuring [Fig. 10(c)], as known from 2D ferrofluids.<sup>358</sup> Long chains<sup>359–361</sup> and lattices<sup>362</sup> form at liquid–liquid and liquid–air interfaces in alternating in-plane magnetic fields where the balance between viscous and magnetic torque, and magnetic attraction and hydrodynamic repulsion govern the stability and mechanical response of the dynamically stable, ordered structures. Jamming these assemblies will inhibit the mobility of individual particles, producing a locked remanent magnetization of the entire droplet. This novel approach stimulates to reimagine magnetism and microrobotics from the perspective of liquids with solid-state functionalities<sup>355</sup> and prospective applications to viscosity engineering, magnetically functionalized liquid–crystalline and plastic–crystalline phases,<sup>363–365</sup> organic synthesis in living cells,<sup>366</sup> and encapsulation and triggered release of cargo.<sup>354,361</sup>

## 2. 3D spin frustration

The expansion of frustrated dipole systems from planar artificial spin ice structures into 3D space will be essential to the resemblance of inherently 3D, frustrated exchange-coupled materials.<sup>367</sup> Although providing valuable insight into spin frustration, the current approach is restricted to 2D planar systems and improperly scaled nearest and next-nearest neighbor interactions due to the finite size of nanoislands. Advances in focused electron beam induced deposition, two-photon lithography and electrochemical deposition will allow for synthesizing single- and multi-component nanostructures where magnetic moments are confined to vertices or connecting segments. This bottom up approach enables a pathway to design 3D geometrically frustrated heterostructures with various symmetry and geometry of isotropic lattices, 2D layered structures, and deliberately disordered systems. Tailoring shape and size (extrinsic) or anisotropy, exchange and Curie temperature (intrinsic) of multi-component heterostructures translates to an ensemble of magnetically harder and softer materials. While single-component nanostructures are simpler to manufacture, they are less realistic since exchange interactions affect thermal spin fluctuations and local ground states. Multi-component materials with different thermal expansion coefficients will open a path to reversibly transform isotropic into anisotropic spin frustrated systems by changing dipole interactions which is particularly impactful for isotropic macrospins with negligible energy barrier, such as XY, Kitaev spins, and spin glasses. The realization of these complex transitions is unique to macrospin incorporated into a nanostructure matrix and not feasible to achieve in a conventional way, e.g., changing temperature and studying systems with various lattice parameters. While the primary

interest in 3D frustrated heterostructures relies on basic science, they may also find application in random number generation or magnetic dipole logic.<sup>368</sup>

## 3. Non-collinear spin textures in 3D nanostructures

The accumulated knowledge of and advances in synthesis, characterization, and numerical modeling of 3D nanostructures will help boost research efforts to manufacture 3D networks for spintronics (race track memory), magnonics (spin wave excitation), and neuromorphic computing (spin oscillator). These prospective applications require high metal purity and minimal interface roughness to guarantee adequate magnetic exchange interactions, and spin-transfer or spin-orbit torque. The current manipulation of spin textures is not only essential with respect to application but also a critical need as structural complexity prevents an effective control by external magnetic fields. We envision two routes going forward addressing domain wall manipulation in 3D networks, and more complex non-collinear spin textures and topological states in nanostructures. There are numerous challenges to overcome before realizing microelectronics based on topological states in 3D networks.

The coherent, synchronized motion of domain walls via spin-transfer or spin-orbit torque necessitates replica of domain walls with the same chirality, type, magnetic moment, width, and thickness. Since domain wall shape and type are governed by extrinsic (shape, thickness, curvature) and intrinsic (exchange, saturation magnetization, anisotropy) properties, particular attention will be devoted to architectures of multiple components, i.e., vertices and connecting segments, bent and modulated nanostructures that affect both kinetics and dynamics of domain walls. To some extent, slightly different domain wall velocities can be compensated using shift registration in the form of periodic pinning sites. Similarly important is to ensure compatibility with planar systems, which demands a mean to effectively couple chiral spin textures into and out of the 3D networks.<sup>337</sup> Alternatively, new ways to nucleate and manipulate chiral domain walls in 3D networks are needed, taking advantage of, e.g., local stray fields predefining domain wall chirality and varying cross section to alter current density beyond the nucleation threshold. A combination of strain and temperature manipulation of local magnetic exchange and anisotropy could enable logical operations in the form of gates and splitters. Even considering the most ideal case of an amorphous metallic network without grain boundary pinning, questions remain about contact resistance, heat dissipation, pinning at corners, and curvature effects. The challenge will be to harness rather than to compensate these effects.

The multitude of physical parameters influencing magnetization vector fields and spin excitations in 3D networks makes the study and optimization of individual components essential, particularly, in view of more complex non-collinear spin textures and topological states. A major milestone is the synthesis of 3D nanostructures with inversion symmetry breaking to stabilize and retain chiral, non-collinear spin textures. The common approach of single-crystals with inversion symmetry-broken unit cells is impossible to achieve with the vast majority of bottom-up nanofabrication techniques, and impractical in view of 3D networks and application. This leaves

curvature, interfaces, and short-range order in amorphous materials. Biaxial 3D networks provide interfacial and curvature-induced inversion symmetry breaking, while enabling spin-orbit torque manipulation. It is unclear, though, how non-collinear spin textures wrapping the magnetic shell would transition at vertices and intersections due to changes of topology. Switching from magnetic shell to magnetic core allows free navigation through the 3D network at the expense of compensated inversion symmetry breaking. These symmetry arguments underline the essence of local variations in curvature, interfaces and short-range order to consolidate a sizable DMI.

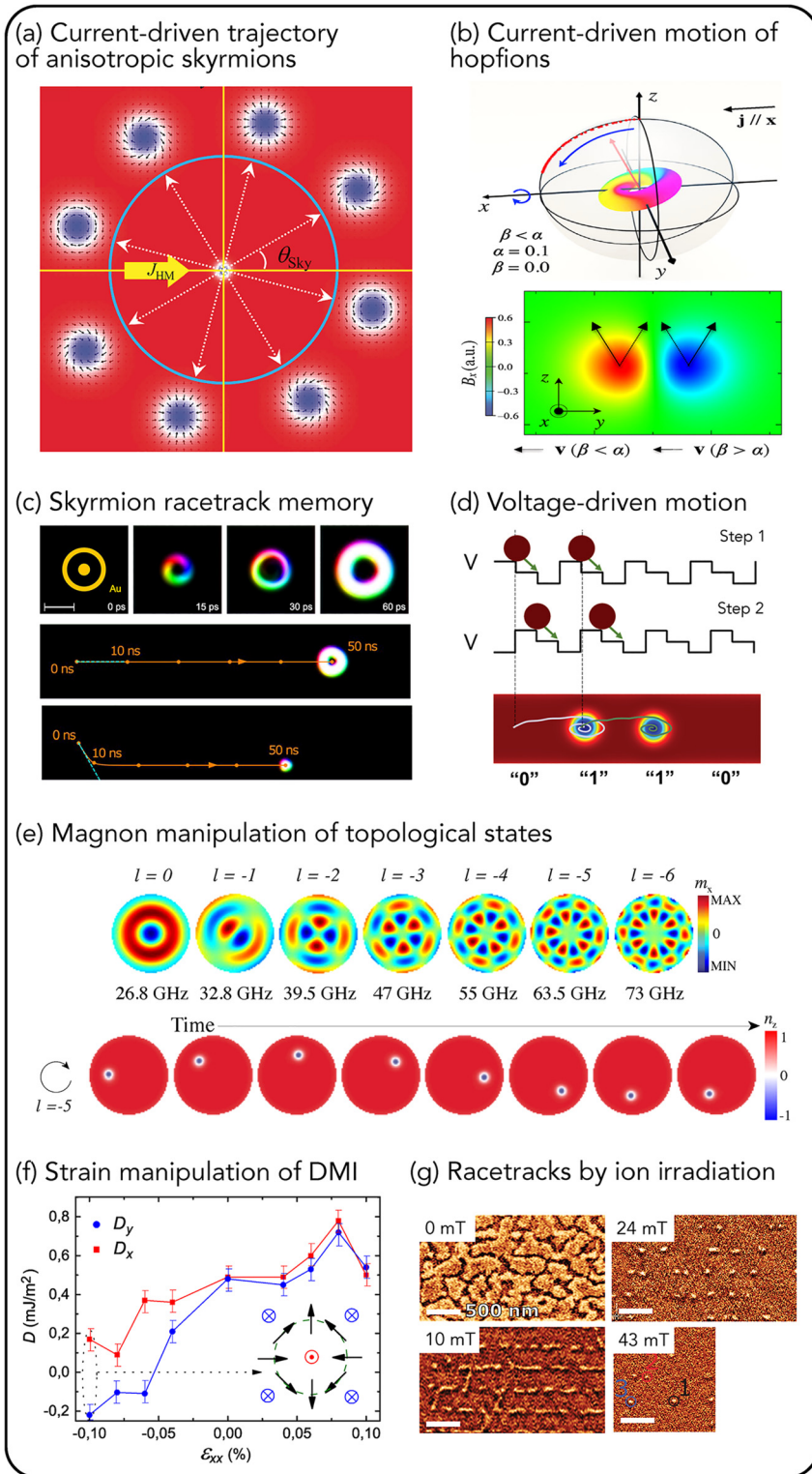
Current efforts to synthesize and investigate cylindrical and tubular nanostructures in view of magnetic states and magnetization reversal process will expand to include domain wall dynamics, spin excitations, and 3D imaging of the magnetization vector field as well as its correlation with local structural and chemical properties in terms of imperfections and engineered chemical/structural segmentation. The latter allows for stabilizing non-collinear spin textures, such as helices, skyrmions and skyrmioniums, which may serve as novel 3D spin oscillators or magnonic crystals for, e.g., speech and pattern recognition. Structural transformation by virtue of thinning (conical shape) or bending (curvature) will further provide means to localize non-collinear spin textures and transition regions between states with distinct topology. This includes reversible switching between collinear and non-collinear spin textures in bent nanostructures, such as helices and rings, harnessing strain-mediated curvature modifications to the magnetic exchange without magnetostriction, magnetic fields, and current flow. While a first demonstration can be given by mechanical stretching and compression, designing artificial magneto-electric materials<sup>63</sup> will involve incorporating nanostructures in, e.g., piezoelectric solgel lead zirconate titanate matrices.<sup>369</sup> 3D magnonic crystals will likely be realized using cylindrical or tubular nanostructures with longitudinal and azimuthal magnetization configurations owing to a profound theoretical understanding and significant advances in synthesis capabilities. The emanation of magnetic spin waves from domain walls separating uniformly magnetized domains simplifies analysis, and addresses the fundamental question of curvature-driven magneto-chirality selection of vortex domain walls and a unidirectional spin wave propagation in tubular geometries that can be tuned by magnetic fields, strain and curvature. A more challenging subject is the spin wave emanating from non-collinear, topological spin textures in 3D nanostructures, and their dependence on both structural and magnetic properties, including, in particular, chirality, topology, and periodicity. This close relationship makes them highly appealing from the perspective of quantum materials for non-volatile, analog information processing.

## B. Topological states stabilized by curvature and short-range order

The research on topological solitons in condensed matter will diversify with a strong emphasis on expanding the zoo of topological magnetization vector fields in homogeneous and inhomogeneous materials and harnessing curvature, disorder, strain, and voltage to tailor type, strength, and inhomogeneity of magnetic exchange interactions on the nano- and atomic scales. Overcoming physical and technological limitations of ferromagnetic Néel and

Bloch type skyrmions will be addressed by exploring ferrimagnetic, antiferromagnetic, and multiferroic isotropic and anisotropic topological states, such as higher-order skyrmions and hopfions. These studies will thrive on multimodal investigations combining magneto-resistance measurements with magnetic imaging of individual and ensembles of topological states with different chirality, topological charge and dimensions. We anticipate similar procedures for dynamic experiments of spin excitations, such as current-, voltage-, and strain-driven motion, nucleation, and spin wave propagation (Fig. 11). Reconstructing thermally stable 3D magnetization vector field with magnetic tomography will provide unambiguous evidence of its topology. Synthesis and experiment will be guided by numerical modeling of the most realistic possible configurations relying on molecular dynamics, *ab initio*, micromagnetic, and Monte Carlo simulations. A more detailed discussion of technological advances is given in Sec. III. Symmetry and order are regarded essential to quantum materials ranging from superconductors to topological insulators to topological magnetic and polar states. Vector spin exchange, i.e., DMI, is an indirect magnetic exchange interaction mediated by conduction electrons of adjacent atoms that reveals, similar to RKKY exchange coupling,<sup>370–372</sup> a spatial oscillatory behavior of both sign and magnitude and is highly sensitive to structural and chemical order.<sup>373</sup> The corresponding local DMI can be homogeneous, inhomogeneous, or random on the microscale and ideally requires a sub-atomically accurate placement of elements and atoms to tailor topological objects and their current-induced motion [Figs. 11(a) and 11(b)].<sup>56,212</sup> However, this is experimentally impractical in view of both efforts and materials synthesis. Instead, we envision that research will focus on structural and chemical disorder in the form of random substitution/intercalation of atoms, rough interfaces, and amorphicity to tailor interatomic exchange on the atomic and nanoscale. Recent theoretical and experimental works have shown promise of these alternate, unconventional means and reinforced the need for a profound understanding of fundamental mechanisms. This ranges from probing and understanding to engineering and harnessing curvature, structural and chemical short-range order in exchange-coupled systems to stabilize topologically non-trivial states with unprecedented small feature sizes and tailored symmetry. Room-temperature skyrmions spanning a few nanometers are currently futuristic but may be realized by enlarging the mean distance between spin-polarized atoms via pseudo-random atom substitution in inversion symmetry-broken systems that leaves the exchange stiffness and DMI exchange constant unaffected.<sup>59</sup>

Amorphous magnetic materials exhibit, due to suppressed electron transfer between transition metal atoms and enlarged local density of states and spin-orbit coupling,<sup>187</sup> increased magneto-resistance and Hall effects and provide greater flexibility in materials synthesis and manipulations via current, voltage, strain, and curvature. In contrast to single-crystals and epitaxial films, they can be grown on virtually any planar, curved or modulated substrate, and will allow for exploring compositions and phases inaccessible in crystalline form due to, e.g., phase segregation. Minimal magneto-crystalline anisotropy and sensitivity to short-range order make amorphous materials ideal prototypical systems to examine curvature-driven DMI, local inversion symmetry breaking and spin chirality selection in terms of stabilized topological magnetic states and magneto-transport phenomena (Fig. 3). This includes, in



**FIG. 11.** Manipulation strategies for topological magnetic states, excluding curvature-induced effects discussed in Fig. 3. (a) Spin Hall effect-induced motion of anisotropic skyrmions. Reproduced with permission from Jin *et al.*, Appl. Phys. Lett. **114**, 192401 (2019). Copyright 2019 AIP Publishing LLC. (b) Current-driven motion of hopfions by spin-transfer torque revealing rotation and expansion or shrinking depending on spin damping  $\alpha$  and non-adiabatic coefficient  $\beta$ . Bottom image shows cross-sectional view of emergent magnetic field with velocities for vortex and antivortex configuration. Reproduced with permission from Liu *et al.*, Phys. Rev. Lett. **124**, 127204 (2020). Copyright 2020 American Physical Society. (c) Optically induced current pulse nucleation and current-driven displacement by spin-orbit torque of skyrmionium as compared with skyrmion. From Göbel *et al.*, Sci. Rep. **9**, 12119 (2019). Copyright 2019 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (d) Voltage control of magnetic anisotropy mediated by adjacent dielectric layer for directional motion of topological states. Reproduced with permission from X. Wang, W. L. Gan, J. C. Martinez, F. N. Tan, M. B. A. Jalil, and W. S. Lew, Nanoscale **10**, 733 (2018). Copyright 2018 Royal Society of Chemistry. (e) Magnetic trapping of magnetic skyrmion by vertical magnons with large orbital angular momentum. Reproduced with permission from Jiang *et al.*, Phys. Rev. Lett. **124**, 217204 (2020). Copyright 2020 American Physical Society. (f) Strain manipulation of magnetic exchange interaction in Co/Pt multilayers. Reproduced with permission from Gusev *et al.*, Phys. Rev. Lett. **124**, 157202 (2020). Copyright 2020 American Physical Society. (g) Formation of skyrmions by helium ion irradiation-induced modifications to magnetic anisotropy and exchange in Pt/Co/MgO. Reproduced with permission from Juge *et al.*, Nano Lett. **21**, 2989 (2021). Copyright 2021 American Chemical Society. (a)–(e) and (f) and (g) are numerical and experimental data, respectively.

particular, the visualization of topological states with different shape, size, and topology, stabilized by isotropic and anisotropic nanoscale indentations and sculptures, and quantification of anisotropic exchange interactions and magneto-transport properties along, e.g., grooves. Selective release from the substrate and mechanical manipulation of free-standing films in the form of local curvature, bending, tensile and compressive strain and pressure are additional intriguing routes to alter magnetic exchange and nucleate and switch topological states. Electric voltage can be used to manipulate magnetic exchange and anisotropy via modifications to the electron density of states near the Fermi level [Fig. 11(d)]<sup>221</sup> and strain-mediated coupling [Fig. 11(f)],<sup>228</sup> altering both degree of inversion symmetry breaking and magneto-resistance. The high sensitivity to short-range order of amorphous materials on the verge between conductors and insulators will enable strain tuning of DMI far exceeding the values for polycrystalline films of one order of magnitude using 0.1% in-plane deformation.<sup>228</sup> The spatial variations in voltage consolidate a reconfigurable curved interface with respect to magnetic exchange that, in principle, can resemble mechanical curvature and induce a local DMI. These efforts can be combined with helium ion irradiation to tailor magnetic exchange, anisotropy, and magnetic moment on the nanoscale [Fig. 11(g)].<sup>218</sup> The latter allows for writing tracks for skyrmion nucleation and motion preventing deflection due to the Magnus force [Fig. 11(c)].<sup>207</sup> Since voltage manipulation of the electronic structures and magnetic exchange is typically limited to insulators and ultra-thin materials due to screening effects in conductors, it will be highly interesting to see experimental studies on the potential of amorphous materials.

Initial investigations will concern extended and nanopatterned homogeneous systems whose compositions are chosen according to their crystalline B20 counterparts with sizable DMI. In long-term, heterogeneous and individually optimized layered structures with continuously varying composition, elements, morphology, and magnetic properties may emerge, enabling, e.g., the formation of topological knots with the unique current-driven motion [Fig. 11(b)].<sup>212</sup> This includes different types and strengths of exchange interactions, magnetic anisotropy, and transition temperatures, as well as multifunctional films with, e.g., magnetic and ferro-/piezoelectric properties. These materials have been synthesized as layered heterostructures or single-crystals and been subject to compatibility limitations. The latter are lifted in amorphous media requiring only short-range order. Multifunctional materials promise voltage, strain, and curvature control of topological magnetism, antiferromagnetism, and multiferroism, magnetic control of polar topological states, and amorphous topological insulators and superconductors<sup>6,374</sup> whose properties can be locally configured by topological magnetic states. An alternate route will focus on atomic layered structures to host topological states spanning a few atoms while taking advantage of negligible damping/pinning due to hybridization of ordered  $p$ -orbitals. Both characteristics make 2D van der Waals materials exceptionally susceptible to disorder and voltage and enable a selective release from the substrate to examine curvature and strain effects all-electrically and via magnetic imaging.<sup>48–51</sup>

Discovering new materials and means to manipulate individual topological states can, in long-term, be accompanied by investigations of collective behavior and spin excitations. This pertains to

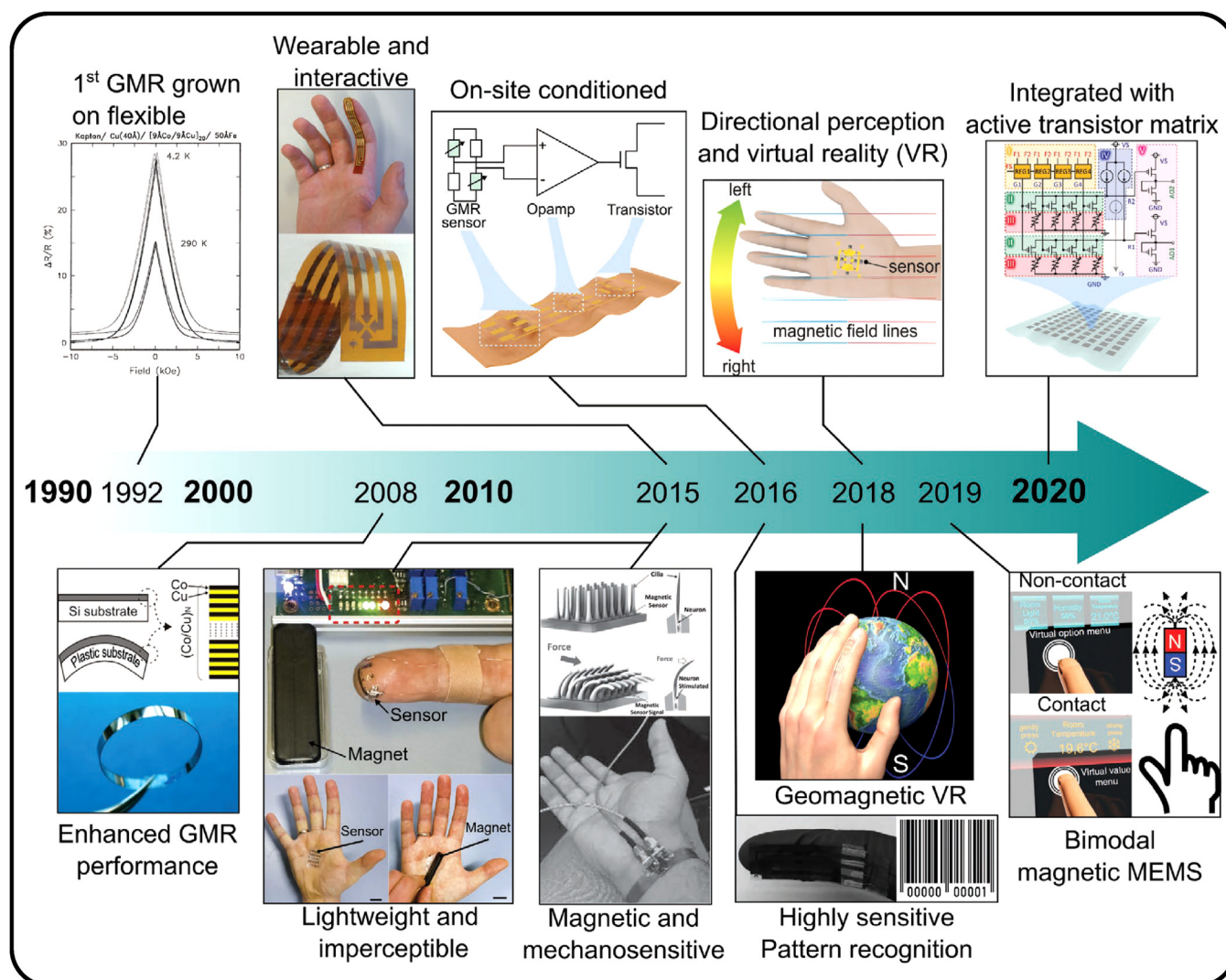
both magnetization dynamics and phase transitions between topologically distinct states, and their relation to structural and chemical short-range order, and local DMI. Spin waves are interesting with respect to disorder-induced topological magnonics,<sup>7</sup> and their lateral and particularly vertical propagation along, e.g., skyrmion tubes, which is critical to envisioning configurable 3D magnonic crystals. Vertical magnons with large orbital angular momentum provide further means to manipulate topological states [Fig. 11(e)].<sup>375</sup> The advantage of designing and implementing curved vector fields over structurally predefined curvature is a new way to tune on-demand the spin wave dispersion, i.e., band structure and topology, through twisting and deforming, or altering the topology of the magnetization configuration. In contrast to topological states confined to 3D nanostructures, extended films offer further collective behavior and potentially a route to design 3D networks of topological states and (topological) spin wave guides.

## V. TECHNOLOGICAL PERSPECTIVE

Despite a strong focus on basic sciences and the early stage of research and development, numerous technological applications of curved magnetic geometries have been proposed and, in part, been realized. They differentiate themselves from planar technologies by enhanced performance, novel functionality, and/or higher efficiency (lower power consumption). Similar to scientific advances, structural properties have taken the lead in both sensing and micro-robotics applications. We anticipate a growing interest in structural, chemical, magnetic, and electronic curved geometries in quantum materials to manipulate chiral and topological states for novel sensing and microelectronics based on spintronics. The fundamental aspects of adding curvature as a critical parameter to magnetic materials were discussed in detail in Secs. II and IV.

### A. Sensing

One of the earliest and most tangible beneficiaries of curved magnetic geometries are flexible and stretchable magneto-resistive sensors (Fig. 12),<sup>376</sup> which can be synthesized by conventional thin film deposition directly onto a flexible and/or stretchable polymeric substrate or via selective release and subsequent transfer onto virtually any surface. The high-quality structural, chemical and magnetic properties enable sensing capabilities, comparable with rigid specimens on, e.g., silicon wafers, relying on giant magnetic resistance and impedance,<sup>153,377,378</sup> Hall effect, and giant stress resistance and impedance.<sup>379</sup> While functional multilayer stacks placed either in the neutral plane or onto micrometer-thick foils experience minimal impact from strain and stress, films on thicker substrates suffer magnetostriction which benefits giant stress impedance measurements in the form of an altered magnetic susceptibility and anisotropy. These characteristics provide the foundation for sensing fluid and gas flow (bending), thermal and mechanical expansions of planar and curved geometries (interfacial strain), and the physical orientation within a constant magnetic field or of a variable magnetic field as an inexpensive and thin alternative to semiconductor Hall probes. The latter will empower contactless position sensing for magnetic bearings, 3D “touch” screen and wearable navigation devices (position and movement in 3D), and on-skin



**FIG. 12.** Development of magnetosensitive e-skins based on magneto-resistive sensing. From G. S. Canon Bermudez and D. Makarov, *Adv. Funct. Mater.* 2007788 (2021). Copyright 2021 Author(s), licensed under a Creative Commons Attribution (CC BY) license.

interactive electronics for, e.g., augmented and virtual reality applications.<sup>376,380–383</sup> Switching from extended films to 3D nanostructures, such as nanopillars, tubes, and helices, will offer spatially resolved sensing capabilities<sup>384–386</sup> on the submicrometer scale and improved sensitivity to both magnetic field and stress. Magneto-resistive vector field sensing could be realized with 3D networks of nanostructures. Given that the magnetic anisotropy and susceptibility of superparamagnetic nanoparticle coatings are highly dependent on the assemblies' short-range order, they can be used to monitor and detect thermal and mechanical expansions as well as cracks in and twists of conducting wires based on impedance changes. Similarly, local magnetic fields in liquids can be probed by analyzing self-assembly of superparamagnetic

nanoparticles. The magnetic phase transition from paramagnetism to ferromagnetism of superparamagnetic nanoparticles upon jamming in liquid environment could be adapted to sensing pH with bio-compatible hydrogels.<sup>387,388</sup> The latter are highly sensitive to small changes in pH leading to a hysteresis-free shrinkage and expansion, which will affect the mean distance between embedded nanoparticles and their remanent magnetization and boost sensitivity.<sup>389,390</sup>

### B. Microrobotics

Magnetic nano- and microstructures dispersed in liquid and gaseous environments are strong contenders for microrobotics



because of a high susceptibility to external magnetic fields, which enables the remote control of the translational, and rotational motion, orientation, and direction, and the selection of different mode of operation. Mechanical actuation and selective transformation of shape-morphing micromachines with distinct local magnetic and elastic properties<sup>294–296</sup> can be realized by chemically, temperature or magnetic field-driven structural deformation, and adapted to complex origami [Figs. 13(a) and 13(b)],<sup>295,297,391</sup> cargo delivery in liquid<sup>293,294,298,354,361</sup> and gaseous<sup>294</sup> environments, viscosity/turbulence engineering (microfluidics),<sup>359,361</sup> and surface roughness/modulation (optics). The magnetic functionality is typically given by ferromagnetic nanoparticles, embedded in elastic films or magnetic films with tailored magnetic properties, and the mechanical response to a rotating or constant magnetic field. The latter offers quasi-static<sup>297</sup> and dynamic [millisecond time scale, Fig. 13(c)]<sup>298</sup> actuation, including microrobots that walk, crawl, roll, and climb in air and liquid environment,<sup>294</sup> bio-inspired flagellated micromachines [Fig. 13(d)],<sup>293</sup> and printing of complex 3D structures that emerge from 2D planar films.<sup>295</sup> These developments pave the way toward prospective applications in life sciences and engineering, such as drug and cargo delivery, directional tissue growth, microsurgery, and artificial fertilization, as well as functional and reconfigurable microfluidic channels, adaptive optical elements, viscosity engineering, local magnetic field sensing and generation, and actuation.<sup>9–11</sup> The reconfigurable magnetization of ferromagnetic liquid droplets provides a mean to promote magnetically aligned cell differentiation and proliferation, which is particularly appealing for blood vessels, cartilage, and nerve tissue regeneration in living cells.<sup>366</sup> In-field assembly and jamming of mixed phases of non-magnetic and superparamagnetic nanoparticles on liquid–liquid interfaces will enable reversible magnetic field-sensitive nanopatterning and designing birefringent, refractive, diffractive and potentially chiral liquid optical components. Choosing ligands with the potential to significantly reduce interfacial tension will provide means to generate micrometer, potentially, sub-micrometer, droplets from parent specimens owing to spontaneous emulsification. A magnetic field promoting the assembly and agglomeration of nanoparticles at the interface will enable a stimulated emulsification in the form of an explosive release of ferromagnetic microdroplets. The viscosity of lubricating liquids can be enhanced by DC magnetic field-induced assembly of superparamagnetic nanoparticles into chains, tubes, flakes, or rings, which decelerates translational and rotational motion of motors; disassembling may occur naturally at remanence or within an AC magnetic field.

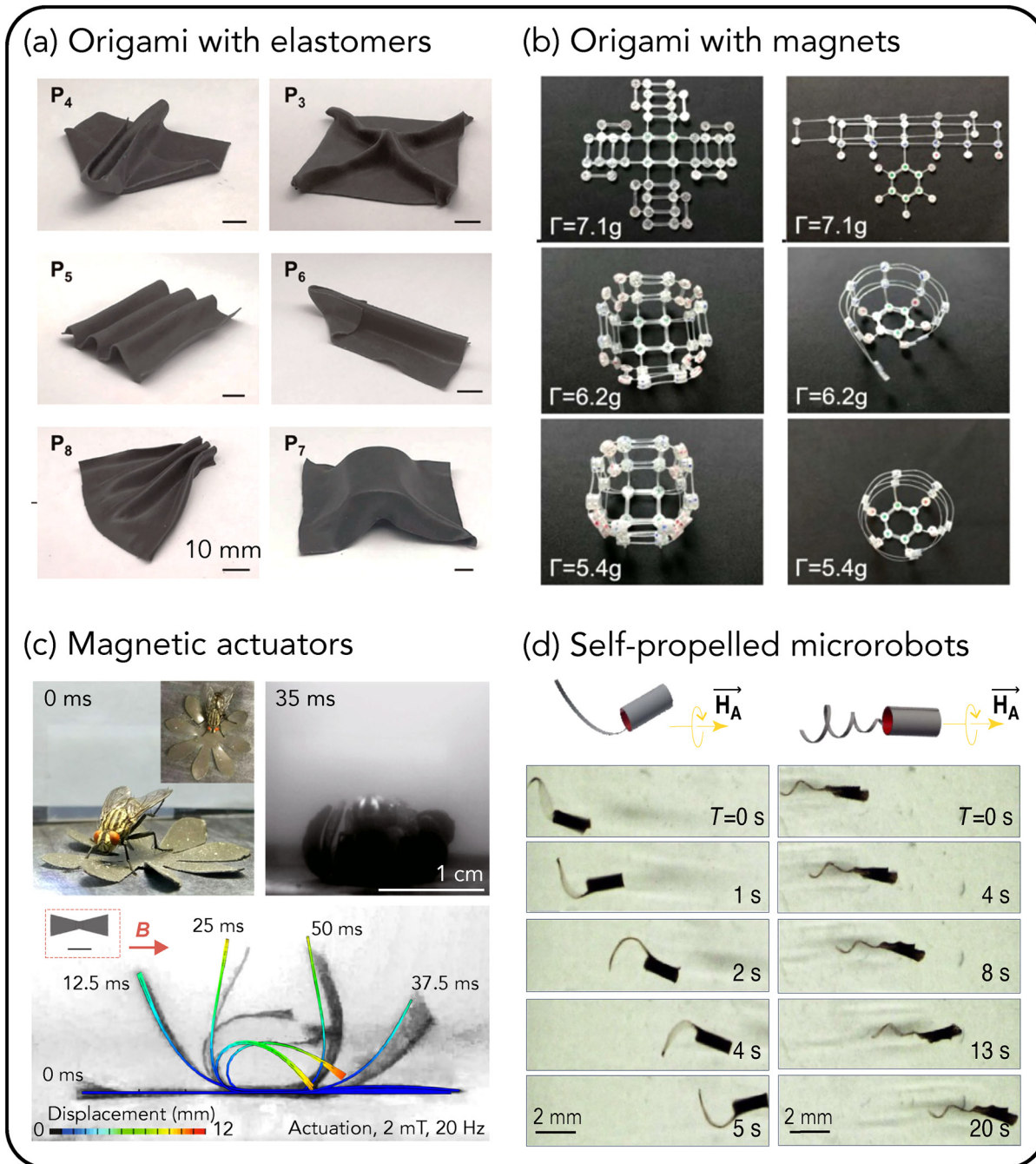
### C. Microelectronics

Compared with sensing and microrobotics, realizing low-power microelectronics by harnessing curved geometries is a long-term effort that requires substantial scientific, technological and, to some extent, conceptual advances regarding, in particular, implementation and integration. Examples range from more conventional mechanisms, such as dipole spin frustration and current manipulation, to voltage, curvature, and topology control. Spin frustration in 3D heterogeneous nanostructures may find application in random number generation and magnetic dipole logic.

Current-driven domain wall manipulation and propagation in 3D networks provide a path toward 3D racetrack memory devices and domain wall logic, as recently demonstrated in 2D,<sup>368</sup> with superior storage density owing to minimal footprint. Incorporating curved nanorods in piezoelectric matrices enables voltage-induced switching between collinear and non-collinear spin textures with distinct magneto-resistance; this approach allows for designing artificial non-volatile magneto-electric materials based on strain-mediated curvature modifications to the magnetic exchange without magnetostriction, magnetic fields, and current flow. Thickness-modulated low-damping materials, such as yttrium iron garnet, or 3D networks thereof can be explored in reference to their capability to serve as tunable ferromagnetic oscillators with multiple independent narrow resonances. These efforts will help launch 5G cellular communication services in the originally intended high-frequency band of (24 ~ 28) GHz to bolster future demand in bandwidth and rate, which is prevented by the currently employed complementary metal–oxide–semiconductor (CMOS) voltage-controlled oscillators.

Novel 3D spin oscillators and magnonic crystals for, e.g., speech and pattern recognition can be realized with non-collinear spin textures, such as chiral domain walls, vortices, helices, and topological states, formed in segments of 3D networks or individual cylindrical nanostructures. The close relation between spin wave excitation and spin chirality, topology, and periodicity as well as structural properties offers a significantly larger parameter space than conventional nanospin oscillators and magnonics based on a uniform magnetization. These devices will take advantage of spatial confinement and directionality of the spin excitation (higher efficiency), and a distinct dispersion relation with potential topological features. Prototypical systems may be based on reconfigurable vortices in non-planar antidot arrays, domain walls pinned at corners of bent planar and 3D nanostructures, and domain walls separating uniformly magnetized domains in cylindrical or tubular nanostructures with longitudinal and azimuthal magnetization configurations. Harnessing chiral spin textures in 3D nanostructures and extended films have the advantage of designing and implementing curved magnetization vector fields over structurally predefined curvature and opens a new way to tune on-demand the spin wave dispersion through twisting and deforming, or altering the topology of the magnetization configuration. Extended films offer further collective behavior and a route to design 3D networks of topological states and topological spin wave guides, which is intriguing from the perspective of novel quantum materials. A potential commercialization demands an all-electric characterization of the spin oscillators and magnonic materials probing the transmitted current signal as a fingerprint of thermal and radio frequency spin wave excitations.

Greater flexibility in materials synthesis of quantum materials and correlated electron systems can be accomplished in the form of amorphous and polycrystalline materials with local inversion symmetry breaking. Focusing on the short-range order instead of global symmetry will allow for designing multifunctional materials which are typically incompatible due to mismatching lattice constants, symmetry and phase segregation. These materials offer voltage, strain and curvature control of topological magnetism, antiferromagnetism and multiferroism, magnetic control of polar topological states, and amorphous topological insulators and superconductors whose properties can be tuned by local decoration with topological magnetic



**FIG. 13.** Shape-morphing magnetic materials in gaseous and liquid environment. (a) Magnetic nanoparticles incorporated into elastomer matrix whose short-range order and magnetic anisotropy can be tuned at elevated temperatures in a magnetic field. Depicted shapes emerge from planar sheet in the presence of a magnetic field. Reproduced with permission from Song *et al.*, *Nano Lett.* **20**, 5185 (2020). Copyright 2020 American Chemical Society. (b) Programmed self-assembly of frames equipped with multipole permanent magnets. From Niu *et al.*, *Proc. Natl. Acad. Sci. U.S.A.* **116**, 24402 (2019). Copyright 2019 Author(s), licensed under a Creative Commons Attribution (CC BY) license. (c) Millisecond actuation based on perpendicular magnetized films exposed to linear AC magnetic field. From Wang *et al.*, *Commun. Mater.* **1**, 67 (2020). Copyright 2020 Author(s), licensed under a Creative Commons Attribution (CC BY) license.<sup>298</sup> (d) Bio-inspired flagellated micromachines with magnetization stemming from embedded aligned ferromagnetic nanoparticles driven by a rotating magnetic field in liquid environment. From Huang *et al.*, *Nat. Commun.* **7**, 12263 (2016). Copyright 2016 Author(s), licensed under a Creative Commons Attribution (CC BY) license.<sup>293</sup>

states. Local voltage applications will provide a mean to consolidate a reconfigurable curved interface with respect to magnetic exchange, and to alter local DMI. The latter is envisioned to promote the formation of complex 3D magnetization vector fields all-electrically, e.g., creating and deleting skyrmioniums and antiskyrmioniums, as well as anisotropic solitons like topological knots, which is essential to post-CMOS microelectronics.

## VI. CONCLUSION

Harnessing the curvature as a design parameter to tailor and manipulate magnetic properties of non-collinear and topological states as well as of 3D magnetic nanostructures is a vital emergent field with ample opportunity for basic and applied sciences. Despite its primary focus on basic sciences and early stage of research and development, a multitude of prospective applications have emerged, including magnetic field and stress sensing, microrobotics, and information processing and storage. Their realization requires an integrated approach of modeling, synthesis, and characterization across multiple length scales. This Perspective presented recent advances in basic and applied sciences and technology in the context of ongoing research efforts that we hope will guide and stimulate future directions.

## AUTHORS' CONTRIBUTIONS

R.S. wrote the manuscript with suggestions from E.Y.T. and P.F.

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## DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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