

# Science and technology of 3D magnetic nanostructures

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For nearly half a century, the era of nanoscience was driven by the paradigm that the reduction in dimensions in nanomaterials would provide a deeper understanding of the fundamental building blocks at the atomic and molecular level, resulting in novel material properties, behavior, and utilization in nanotechnologies. Specifically, for magnetic materials, this triggered enormous research efforts in spintronics and magnetic nanostructures. However, about ten years ago, it was realized that the extension of accomplishments from nanoscience and nanotechnology into the third-dimension will not only open new opportunities in magnetic materials<sup>1,2</sup> due to additional levels of complexity or phenomena that can only exist in 3D, such as chirality, but will also yield substantial challenges for the synthesis, theory, and characterization of such artificially designed 3D systems. Rapid improvements in fabrication technologies,<sup>3–10</sup> theories predicting curvature-driven novel energy terms,<sup>11–13</sup> and new types of spin-textures that stabilize due to geometrical effects, unique topology,<sup>14</sup> and frustration,<sup>15</sup> as well as new experimental approaches to validate 3D spin textures and their behavior emerged. With the development of nanoscale magnetic imaging techniques<sup>16–19</sup> that can be characterized even quantitatively, the complete 3D magnetization vector, with a precision down to magnetically relevant lengths and timescales, has helped elucidate the impact of nanoscale curvature<sup>19–21</sup> on well-known spin textures as well as demonstrate the existence of new topological spin systems such as Bloch points,<sup>22</sup> merons,<sup>23</sup> and Hopfions.<sup>24</sup>

One of the initial technology drivers toward 3D magnetic nanostructures was pushed by a desire to extend magnetic storage into the third dimension, by harnessing domain wall motion through magnetic nanowires that rise out of the sample plane, which would naturally increase the storage capacity at reduced areal density,

promising greater performance and energy efficiency.<sup>25</sup> Despite the fact that racetrack technology might still have a long way to go before entering the market, this initial field has triggered numerous efforts toward complex 3D magnetic nanostructures demonstrating a range of scientifically exciting and potentially technologically relevant phenomena, including novel ways to control static and dynamic domain wall behavior,<sup>26</sup> emergent physics in topological and chiral 3D spin textures, frustration in spin ice systems on nanostructured lattices,<sup>15,27,28</sup> and controlled magnonic transport.<sup>29</sup> With a crescendo in the field approaching, we present a collection of articles that represent a cross section of activity in 3D nanomagnetism, while reflecting upon the challenges that lie ahead.

It was in 2008 when the first magnetic racetrack memory was proposed at IBM Almaden Research Center by Parkin *et al.*<sup>25</sup> While it was clear that the technology was a true shift in paradigm for data storage, many challenges lay ahead with respect to how domain walls could be written, propagated, and detected. The key concept of this design was the 3D nature of magnetic nanowires that harbored domain walls as information carriers. Understanding the relevant domain wall topology, dynamics, and ways in which such walls pin is important in developing future racetrack technologies. Fernandez-Roldan and Chubykalo-Fesenko investigated the propagation of Bloch point domain walls (BPW) under a spin-polarized current.<sup>30</sup> As previously noted,<sup>31</sup> the generated Oersted field is found to have a profound impact upon domain wall (DW) dynamics and pinning. When the Oersted field has the same chirality, as the outer wall spin texture, it is found to propagate at ~300 m/s without dynamic instabilities. When the chirality of the wall structure and the Oersted field are opposed, the velocity is found to be greatly reduced and, in some cases, the wall is even pinned. The work

highlights the importance of Oersted fields in magnetic racetracks based on cylindrical nanowires. Askey *et al.* used micromagnetic simulations in order to investigate in detail how DW topology varies in cylindrical Ni nanowires.<sup>32</sup> In the diameter range of 100–240 nm, two distinct topologies have previously been observed: that of the BPW wall with a singularity in the center and that of the asymmetric-transverse domain wall, containing surface vortices and an internal vortex tube. Both walls are reproduced in the phase diagram determined by Askey *et al.*, but, surprisingly, a third metastable wall is found with both surface vortices and two internal Bloch points. Upon application of a field, this new dual BPW wall is found to transform into a conventional BPW, via ejection of a single Bloch point and annihilation of surface vortices. The work suggests that the metastability of DW topology needs to be investigated carefully before technologies based upon cylindrical wires can be considered.

Cylindrical nanowires with a large diameter and magnetization have previously been found to host novel chiral states whereby the magnetization curls around the wire circumference.<sup>33</sup> Brajuskovic *et al.* studied the remanent state and magnetization reversal of NiFe cylindrical nanowires by using micromagnetic simulations and Lorentz Transmission Electron Microscopy (TEM).<sup>34</sup> Their focus is upon the simulation and measurement of 150 nm diameter nanowires and upon field-driven reversal from a uniform state. These are found to host a “double-helix” spin configuration whereby two helices wind around the nanowire diameter, similar to states seen previously in 3D nano-printed double helix geometries.<sup>14,35</sup> The two magnetic helices have opposite magnetization components along the nanowire direction. The study suggests that there may be a diverse range of chiral states in larger diameter soft magnetic nanowires. Patterning chiral magnets with intrinsic Dzyaloshinskii–Moriya interaction (DMI) into cylindrical nanostructures is also of recent interest and can be used to realize a variety of magnetic textures. Savchenko *et al.* carried out micromagnetic simulations and off-axis electron holography of FeGe nanocylinders.<sup>36</sup> A key experimental observation is that of a dipole string (toron), a configuration that is comprised of two Bloch points with opposite topological charge. Interestingly, the damage introduced during the focused ion beam (FIB) process is found to be the key in the stabilization of the texture.

Magnetic random access memory (MRAM) is a technology based upon arrays of magnetic tunnel junctions (MTJs), with spin transfer torque being used to manipulate the magnetization within the free layer of individual cells. Although the initial hopes that MRAM would transform magnetic technologies at large seem to have not yet materialized, there are already several realizations of MRAM technologies occupying certain niches in the market. An increase in areal density requires a reduction in the size of individual MTJs while maintaining thermal stability. One means to do this is to pattern the MTJs into cylindrical geometries and harness a double tunnel barrier scheme, as investigated in the early 2000s. A second scheme relies on an elongated free-layer, where an increased thickness provides stability via shape-induced perpendicular anisotropy. These complex multilayer nanoscale systems often have unknown magnetic behavior within individual layers, making device optimization difficult. An effective means to study the magnetism in such devices is outlined by Almeida *et al.*, who presented a method whereby individual rows of elements are transferred from large-scale arrays and imaged using electron holography.<sup>37</sup>

*In situ* heating allows the study of thermal stability within individual elements.

The field of artificial spin-ice (ASI)<sup>38,39</sup> is another scientifically exciting topic involving the placement of interacting single domain magnetic nanowires into frustrated configurations, allowing exploration of ground state ordering and emergent excitations. Originally limited to 2D spin arrangements in symmetric and asymmetric lattices, the extension toward 3D ASIs will provide unprecedented insight into the phenomenon of frustration and associated spin dynamics. To date, the field has been dominated by planar geometries whereby nanowires are placed into, e.g., square<sup>39</sup> or Kagome<sup>40,41</sup> geometries in an attempt to capture the aspects of spin geometry in bulk pyrochlores. The study of such simple structures and beyond has become a vibrant field allowing realization of idealized models in statistical physics,<sup>42</sup> topological frustration,<sup>43</sup> and emergence.<sup>38</sup> A key disadvantage with 2D systems is that they cannot capture the energetics of bulk systems, and this is simply because one cannot arrange four spins on a 2D plane in such a way that every spin makes the same angle with nearest neighbors. Moving artificial spin-ice to the third dimension allows one to surmount this problem and place magnetic nanowires in a diamond-bond lattice. This feat has already been achieved with large-scale diamond-bond 3DASI lattices<sup>27</sup> showing magnetic monopole propagation upon the surface<sup>15</sup> and exotic charge crystal ordering. A problem that remains in the field is that of imaging the magnetism in large-scale 3D lattices. A first step in this direction is presented by Pip *et al.*, who carried out two-photon lithography and deposition of GdCo in order to realize a tripod structure consisting of three magnetic nanowires.<sup>44</sup> Cutting-edge x-ray magnetic laminography upon the 3DASI building block and subsequent reconstruction shows a low energy 2-in/1-out configuration. The presence of a sheet-film close to the 3D structure can potentially be exploited to control the degeneracy of the states. Overall, the study provides a foundation for imaging 3DASI systems. The macroscopic degeneracy of ASI can also be exploited to produce tunable magnonic devices. Here, the simplest means to exploit the third spatial dimension is to grow an ASI lattice upon a thin film, to tune the modes present in the structure. Negrello *et al.* did exactly this, patterning a NiFe square ASI upon a continuous NiFe thin film.<sup>45</sup> The spin-wave dispersion in a Damon–Eshbach configuration is measured using Brillouin light scattering, and modes are found that are independent of the film, while others are strongly coupled with the underlayer. Overall, dynamic mode coupling in an out-of-plane direction is found to modulate spin wave propagation. A more complex magnonic system based upon a 3DASI is the buckyball. This system is studied via simulations by Cheenikundil *et al.*, who explored the high-frequency modes found within tubular and solid nanowires.<sup>46</sup> The demagnetized system, consisting of only ice-rule vertices, is found to harbor five modes, which are divided into those localized at vertices and standing wave modes across the nanowires. Application of magnetic field increments is found to significantly alter the remanent excitation spectrum, paving the way to tunable magnonic systems.

The propagation, control, and detection of spin-waves<sup>29</sup> provide the basis for new computing<sup>47</sup> and communication technologies whereby signals can be transported over a wide range of frequencies (1–100 GHz) and with no conventional Joule heating. Furthermore, the realization of periodic structures allows systems to be realized with controlled transmission, in analogy to

photonics crystals. Moving spin wave and magnonic structures into non-planar geometries allows such concepts to follow current trends in CMOS, moving circuits to three-dimensions, while also offering new means to generate and control spin-waves. Here, curvature is again expected to be an important geometric parameter, allowing spin-wave non-reciprocity while maintaining key material properties such as damping constant. In order to realize complex magnonic circuits in 3D, direct-write fabrication methods such as two-photon lithography<sup>48</sup> or focused electron beam induced deposition (FEBID)<sup>3,8,49</sup> are required. In both cases, a proof-of-principle is needed to show that materials grown in this way can be integrated with the relevant 2D excitation and detection circuits. Küss *et al.* studied the interaction between surface acoustic waves and spin-waves by coupling planar interdigitated transducers (IDTs) with magnetic microwires grown using FEBID and focused ion beam technologies.<sup>50</sup> A key advantage is that the transmission characteristics of the IDTs can be used to address specific micro-strips while forward volume spin-waves are probed. The study demonstrates that 3D direct-write technologies can be coupled with planar surface acoustics, to produce simple spin-wave systems.

Topological magnetic solitons, where magnetic skyrmions<sup>51</sup> are probably the most prominent examples of two-dimensional spin textures, are seeing a recent increase toward 3D counterparts, such as Hopfions,<sup>24</sup> bobbars,<sup>52</sup> strings,<sup>53</sup> and torons.<sup>54</sup> They require often an inherent symmetry breaking and can be stabilized in multilayer systems with tuned Dzyaloshinskii-Moriya interaction. In particular, Hopfions, which have only recently been observed experimentally in multilayer systems,<sup>55</sup> show exceptional promise in next generation racetrack devices, where they might overcome the unavoidable topological Hall effect seen with conventional skyrmions, which pushes them to the edges of the racetrack. Novel topological spin textures also provide a means of controlling spin waves via their intrinsic configuration. Kotus *et al.* used micromagnetic simulations to investigate spin-wave scattering in a hybrid system consisting of conventional NiFe planar waveguide and a Co nanodot placed at the center, 2 nm above the surface.<sup>56</sup> Magnetostatics perturbs the spin texture of the waveguide, producing an imprint of the skyrmionic configuration. Specific spin-wave excitations are then selected from the dispersion relation, and the impact of the skyrmion imprint is investigated. Three regimes were identified, whereby either modes were preserved in transmission and, in reflection, scattered into alternative modes, or all modes were scattered yielding local interference. Overall, the study suggests that the 3D placement of complex spin textures can be used to tune spin-wave propagation. Skyrmions can also be stabilized in 3D lattices and the periodic potential exploited to produce standing chiral spin waves. Savchenko *et al.* studied this scenario within a theoretical setting.<sup>57</sup> A classical spin model with exchange and DMI terms is utilized to study a thin film geometry with periodic boundary conditions along the x-y directions. Upon in-plane excitation, a series of modes is found, whereby maxima are located at the skyrmion core or between skyrmions. At resonance, the standing waves take a chiral character with helical modulation due to competition between exchange and DMI. An interesting idea with respect to the identification of topological spin textures is to use spin-wave spectra rather than a direct imaging via microscopies. Recent work has suggested that such a measurement can be used to distinguish between skyrmions and

Hopfions.<sup>58</sup> Sobucki *et al.* utilized frequency-domain finite element simulations to investigate the spectral signature of Hopfions within a ferromagnetic dot of 200 nm diameter and 70 nm thickness.<sup>59</sup> Two groups of modes were found to be associated with the Hopfion, originating from the outer and inner regions of the spin-texture. The findings suggest it may be possible to identify Hopfions via spin-wave spectra measurements. Although Hopfions have been found to be stabilized in magnetic multilayers, their presence in bulk solids has not yet been demonstrated. However, a new theoretical investigation by Rybakov *et al.* suggests that Hopfions can be found in solids within the framework of a classical Heisenberg model with competing exchange.<sup>60</sup> A parameter space for finding Hopfions in solids is identified, and the calculated magnetic phase, as measured by off-axis electron holography, suggests that detection should be possible.

Another type of magnetic soliton with a 3D spin texture is the chiral bobber that can be derived from a skyrmion tube decreasing in size and terminating in a Bloch point. Evidence of such topological structures has been confirmed experimentally using electron holography, but complete distinction between chiral bobbars, skyrmion tubes, and torons is an exceptionally challenging task with conventional magnetic imaging alone. Again, the spectral fingerprint of such topological spin textures can be a useful means of identification. Bassirian *et al.* utilized micromagnetic simulations to describe a new topological spin texture consisting of two antiferromagnetically coupled Néel bobbars (bi-bobber).<sup>61</sup> Interestingly, the bi-bobber stability is found to be dependent upon the underlying chiralities. After studying static properties, an alternating external magnetic field is utilized to study the breathing modes in the bi-bobber structures, and this was compared to modes seen in skyrmion tubes. The dispersion relations are found to be distinct, again opening up new means to distinguish between similar topological spin textures.

Two further 3D magnetic solitons, that of a hybrid skyrmion tube and 3D chiral droplet, are discussed by Kuchkin and Kiselev in the context of bulk chiral magnets.<sup>62</sup> A novel means of obtaining these spin textures is presented, whereby 2D textures of the same homotopy class are stacked, and by determining the minimum energy path, new stable 3D textures are produced. A phase diagram of magnetic field and magnetic anisotropy is provided to show the regions of stability for the 3D chiral droplet. An alternative means to stabilize conventional skyrmion tubes is using exchange-spring heterostructures,<sup>63</sup> consisting of a soft ferromagnetic layer coupled to a perpendicular magnetized layer, within a cylindrical geometry. Charilaou showed that, when studying such systems using micromagnetic simulations, an external field can be used to reverse the magnetization in the soft layer, yielding a skyrmion tube and a pair of Bloch points.<sup>64</sup> The removal of the field drives the Bloch point toward the open end of the cylinder at speeds of  $\sim 1$  km/s, due to exchange coupling with the hard perpendicular layer. Here, the Bloch point propagation yields high solenoidal electric fields of order  $10^5$  V/m, a response that can be harnessed to produce electromagnetic pulses, similarly to previous predictions.<sup>65</sup>

The topic of 3D nanomagnetism has become a flourishing field within condensed matter physics, which expands the enormous scientific and technological achievements of low dimensional nanomagnetism into the third dimension. The excitement and promises that this new path entails will justify the many challenges ahead with

regard to pushing the boundaries of established ways to fabricate, measure, and model requisite structures and control and understand their underlying magnetism. We hope this special issue will inspire both established and new researchers within the field and beyond.

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## AUTHOR DECLARATIONS

### Author Contributions

**S. Ladak:** Writing – original draft (equal); Writing – review & editing (equal). **A. Fernández-Pacheco:** Writing – original draft (equal); Writing – review & editing (equal). **P. Fischer:** Writing – original draft (equal); Writing – review & editing (equal).

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