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ABSTRACT

The scientific and technological exploration of three-dimensional magnetic nanostructures is an emerging research field that opens the path to exciting novel physical phenomena, originating from the increased complexity in spin textures, topology, and frustration in three dimensions. One can also anticipate a tremendous potential for novel applications with those systems in a magnetic sensor and information processing technologies in terms of improved energy efficiency, processing speed, functionalities, and miniaturization of future spintronic devices. These three-dimensional structures are distinct from traditional bulk systems as they harness the scientific achievements of nanomagnetism, which aimed at lowering the dimensions down to the atomic scale, but expand those now in a tailored and designed way into the third dimension. This research update provides an overview of the scientific challenges and recent progress with regard to advances in synthesis approaches and state-of-the-art nanoscale characterization techniques that are prerequisite to understand, realize, and control the properties, behavior, and functionalities of three-dimensional magnetic nanostructures.

I. INTRODUCTION

One of the hallmarks of nanoscience and its associated nanotechnology was to explore and discover new materials by lowering the dimensionality. Nanomagnetism is the branch in nanoscience that aims to understand magnetic properties and behavior of magnetic materials down to fundamental magnetic length and time scales, which reach deep into the subnanometer spatial and femtosecond temporal regimes. At those scales, proximity and confinement are underlying concepts that enabled the discovery of new phenomena, which has led to applications that are commonly described by spintronics.

Among the most prominent examples is the Giant Magneto-Resistance (GMR) effect, which was discovered as a transport phenomenon in tailored metallic multilayered systems, where the change in the relative orientation of the magnetization, i.e., the coupling of few nanometer thin magnetic films, leads to large changes in electrical resistance due to spin dependent scattering. This effect immediately revolutionized existing technologies, most notably in high density magnetic storage media, where the Tbit/in.^{2} device has become now a commodity in every-day computing applications. The importance of the GMR effect was recognized with the Nobel Prize in Physics to Albert Fert and Peter Gruenberg in 2007, only a few years after they published their foundational experimental observation in 1988[1] and 1989,[2] respectively.

Despite the huge success of spintronics, there is the conceptual question as to whether this approach to lowering dimensionalities can continue or whether fundamental limits emerge that cannot be overcome both scientifically and technologically.

The level and also diversity of scientific and associated technology achievements with research on low-dimensional nanomagnetism can be illustrated by the following examples, which by no

means are meant to be exhaustive. Numerous reviews and monographs on the fundamental physics and applications in nanomagnetism and spintronics are widely available, which we recommend to consult for that purpose.[3–9]

Zero dimensional (0D) magnetic systems, such as colloidal ZnO doped diluted magnetic semiconductor quantum dots, can be readily synthesized[10] and exhibit, e.g., giant Zeeman splitting and high-Tc ferromagnetism that could be useful of ferromagnetic semiconductor architectures in spintronics applications.

Magnetic core shell nanoparticles (NPs) (Fig. 1 top) that are, e.g., composed of a highly magnetic core material surrounded by a thin shell of desired polymer or metal oxide are investigated as

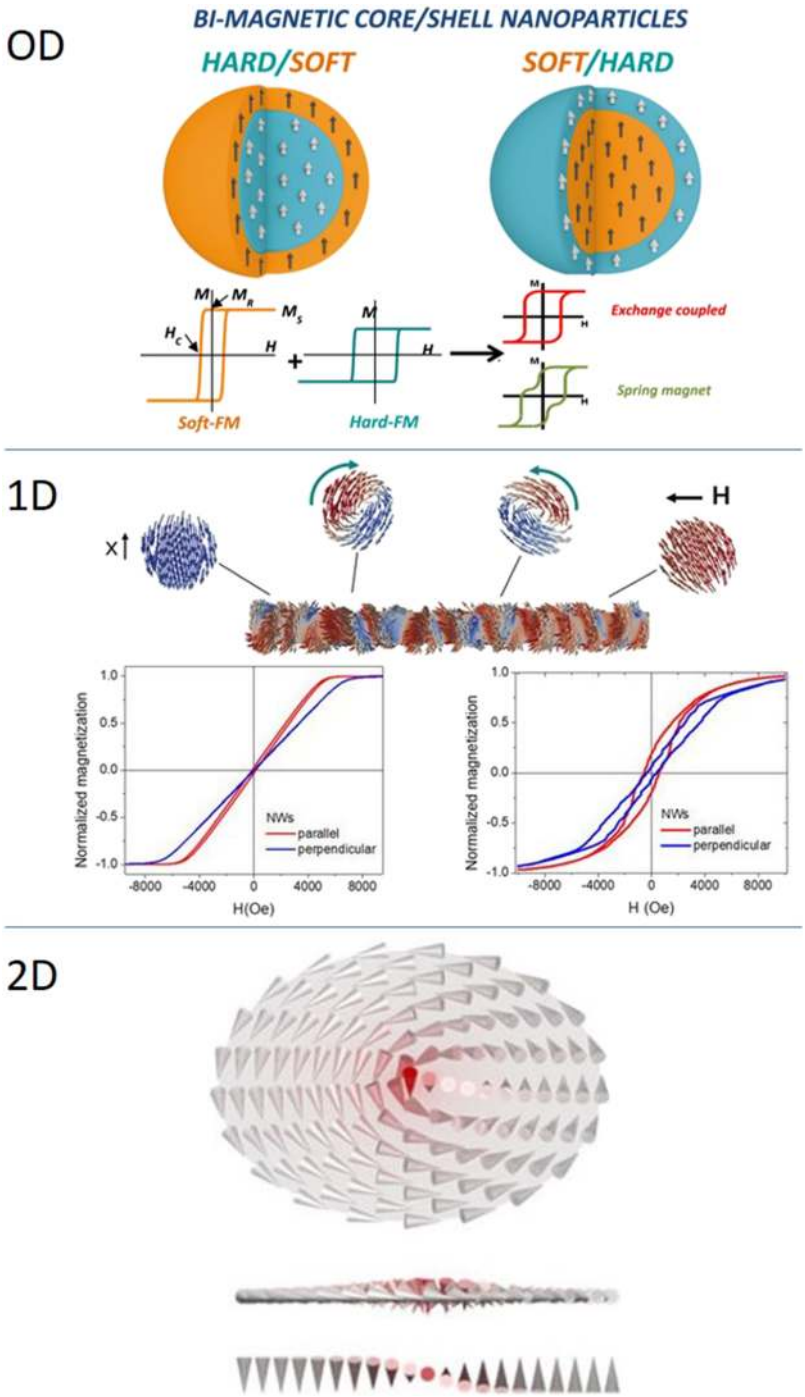

FIG. 1. Spin configurations in various low-dimensional systems. Top: Bimagnetic core/shell nanoparticles (from Ref. 13). Center: Simulated magnetization configuration in a magnetic nanowire (from Ref. 25). Bottom: Schematic drawing of the spin configuration in a magnetic vortex (from Ref. 186).

interesting candidates for biomedical research, such as tissue imaging, drug delivery, or therapeutics.[11,12]

Exchange coupled bimagnetic hard/soft and soft/hard ferromagnetic core/shell nanoparticles exhibit interesting static and dynamic properties that can find wide use, ranging from novel permanent magnets to recording media, microwave absorption, and biomedical applications.[13,14]

Spin torque nanooscillators (STNOs) are nanoscale point contact devices[15,16] that are discussed as future candidates for applications in microwave technologies over a wide frequency range from a few megahertz to tens of gigahertz.[17,18] They use the spin transfer torque (STT) effect, where a spin-polarized current can transfer angular momentum to local magnetic moments of a ferromagnetic system leading to the switching of the magnetization. They also offer an interesting platform to study nonlinear spin dynamics,[19–21] and they are intensely studied as building blocks for future neuromorphic computing concepts.[22,23]

The microscopic spin textures in those 0D magnetic nanostructures are quite often the key to understand the underlying processes that give rise to the observed phenomena in those systems.

Among **one dimensional (1D)** magnetic nanostructures are magnetic nanowires (NWs) (Fig. 1 center) and nanotubes (NTs).[24,25] NWs are extended magnetic systems, where the length is typically orders of magnitude larger than its cross section, e.g., a several micrometer long ferromagnetic system with a disk-shaped cross section of a few nanometer only. Those systems are commonly synthesized through various chemical deposition techniques, e.g., by electroplating using nanoporous templates,[26,27] atomic layer deposition (ALD),[28] or focused electron beam induced deposition (FEBID).[29–31] NWs and NTs are investigated not only as single component materials, such as metallic nanowires, but also as heterostructured NWs and NTs, where, e.g., the composition changes either in segments along the length such as in multilayers or along the radius of the NW/NT to obtain an elongated coated or core-shell arrangement. Compared to 0D nanoparticles discussed in the previous paragraph, the 1D shape of NWs and NTs adds a significant shape anisotropy, which is—amongst other applications—of interest for biomedicine and for increasing the energy product in permanent magnets or in microwave technologies.[32–34]

A different concept for synthesizing 1D magnetic nanostructures are lithographically defined magnetic nanostrips on a surface, e.g., on Si. Their geometry differs from the above-mentioned NWs as the cross section is in most cases a flat rectangle with the thickness being in the few nanometer regime and the width typically a few tens to hundreds of nanometer. They have received significant attention and sparked intense research activity within the nanomagnetism community as a platform for the concept of a so-called magnetic racetrack memory[35,36] or for magnetic logic elements,[37] where magnetic domain walls serve as digital information units that can be moved, e.g., by spin polarized current pulses utilizing the STT effect.[38–44] In all those 1D magnetic nanostructures, domain walls and their internal spin configuration are one of the key components for a fundamental understanding of their properties and behavior.

Two dimensional (2D) magnetic nanostructures are characterized by a confinement of the spins along two directions. A prototypical example is the magnetic vortex (MV) structure[45] that occurs in a spatially confined disk where a certain aspect ratio of disk diameter to disk thickness allows for the existence of nontrivial, i.e., nonsingle, domain spin textures[46,47] (Fig. 1 bottom). The vortex structure is energetically favored as the magnetostatic energy of the single domain state increases with larger disk diameters. As a result, the spin texture of a MV consists of a circulating in plane magnetization that follows the circular shape of the disk thus avoiding magnetic charges. However, closer to the center, where the proximity of neighboring spins would require an antiparallel alignment, the exchange interaction of neighboring spins overcomes this shape anisotropy and pulls the spins out of plane, leading to the core of the MV pointing perpendicularly out of the plane of the disk. With the in-plane magnetization circulating either clockwise or counterclockwise and the out-of-plane magnetization at the core pointing up or down, such a magnetic disk has four degenerate ground states, which made the vortex structure an interesting research object both for fundamental and applied reasons.[45,48–63]

Recently, the role of topology has become a dominating theme in the study of and search for complex states in condensed matter.[64,65] Magnetic skyrmions are the most prominent example, where a nontrivial topology leads to stability against transformation into trivial states, e.g., the ferromagnetic ground state. The interplay of spin-orbit coupling and low dimensionality[66] combined with an asymmetric exchange interaction, the so-called Dzyaloshinskii–Moriya interaction (DMI),[67] that occurs through inversion symmetry breaking at, e.g., interfaces, enables the formation of skyrmions in thin films.

Skyrmion states are classified using the skyrmion number or topological charge $S = \frac{1}{4\pi}\int n(x,y)dxdy$, where $n(x,y) = \vec{m}\cdot(\partial_x\vec{m} \times \partial_y\vec{m})$ being the winding of the magnetization across the soliton. This terminology presumes a 2D spin texture with a depth-independent profile along the z axis. It follows that for a skyrmion $S = 1$ and for a MV $S = \frac{1}{2}$, i.e., a MV can be considered a half skyrmion. Although the first skyrmions were experimentally discovered at low temperatures in bulk materials that exhibit inherent inversion symmetry breaking[68–70] following theoretical predictions,[71–78] the occurrence of skyrmions in thin films as a result of interfacial DMI is now well established.[79–83] Therefore, the exceptional properties of skyrmions with regard to their stability, nanometer size, and dynamics have raised large interest as potential use for novel spintronics logic and memory devices with commonly used thin film multilayered materials, and a plethora of studies has been devoted since to understand and control the creation, detection, manipulation, and deletion of skyrmions using magnetic fields, currents, and voltages. Several recent reviews highlight the achievements and future opportunities with skyrmions and their applications in skyrmionics.[84–89]

Since the discovery of graphene in 2004,[90] which is a strictly two dimensional materials, van-der-Waals heterostructures have become a strong focus of interest[91,92] as they provide a unique platform to combine atomic layers of nearly any material without any constraints for matching lattices that exhibit a wealth of novel properties, including metals, semiconductors, insulators, and superconductors that have high potential to lead to new application and technologies. **Two dimensional** ferromagnetism has been observed in van-der-Waals crystals,[93] and quite recently, even skyrmions were found in such systems.[94]

Beyond **topology** as a guiding principle to discover novel spin textures and magnetic materials, it is the phenomenon of

frustration in a spin system that provides a rich field for scientific discoveries. Among those are spin ices, where symmetry prevents the system to satisfy simultaneously certain ground state configuration of pairwise spins thus leading to frustration and consequently thermal spin fluctuations. Whereas there are a few naturally occurring systems, e.g., pyrochlores, e.g., $Dy_2Ti_2O_7$,[95] it was the ability to artificially design deliberate spin ice systems by using lithographical techniques to arrange two dimensional nanostructures (nanoislands) into tailored lattices,[96,97] e.g., square, kagome, honeycomb, shakti, etc.,[98] to generate geometrical frustration. Each of the individual nanoisland in such a lattice is considered to have a single one-dimensional macrospin, where shape anisotropy determines largely its direction. At the vertices within those lattices, certain spin ice rules have to be applied and can lead to exotic phenomena, e.g., the observation of magnetic monopoles and Dirac strings.[99]

Following those 0D, 1D, and 2D examples, it seems only natural to move into the **third dimension** with magnetic nanostructures. Although, a lot of phenomena in the low dimensional systems mentioned above can be explained by their low dimensional geometry, a profound understanding has to take into account the full three-dimensional arrangement at the subnanoscale. This goes beyond the trivial fact that the spin as a vector quantity has inherently three dimensions. It is the emerging field of 3D nanomagnetism that due to recent developments in instrumentation and tools is able to respond and explore those new research directions in an unprecedented way. This review will provide an update on the scientific opportunities and experimental and theoretical challenges in 3D magnetic nanostructures.

A magnetic nanoparticle (NP) typically consists of hundreds of atoms. To describe the switching of the magnetization of a nanoparticle, one has to take into account the full 3D spin arrangement. Recent developments in high spatial resolution tomographic imaging with aberration corrected transmission electron microscopy allowed for deciphering the 3D arrangement of 23 000 atoms inside a nanoparticle,[100] which have opened a path to expand those capabilities into magnetic nanoparticles. Elemental specificity to study multicomponent nanoparticles, such as core-shell and composite materials, can be provided by magnetic circular dichroism primarily with x-rays[101–105] and also with electrons.[106–108] Electron[109–113] and x-ray[114,115] vector field tomography for magnetic materials are fundamentally different offering sensitivity to the magnetic induction and the magnetization vector fields, respectively.

Other examples to showcase the importance of all three dimensions are boundaries between homogeneously magnetized regions, i.e., domain walls, magnetization switching, and 3D noncollinear spin textures, e.g., topological magnetization vector fields. This recent recognition is contingent with the evolving field of spintronics, relying on spin-transfer torque and topological Hall effect to manipulate and detect chiral spin textures. All-electric manipulation of magnetic domain walls in the form of nucleation, displacement, and transformation strongly depends on their internal structure, e.g., width, chirality, and symmetry (Bloch or Neel type), including depth dependence of the magnetization. An optimization of magnetic and magneto-transport properties hence necessitates knowledge about 3D arrangement of spins at the subnanoscale, pushing experimental limits in view of sensitivity and spatial resolution of state-of-the-art instrumentation.

For example, it has been shown both experimentally and in simulations that in thick films, the domain wall spin textures deviate from a basic symmetric Bloch or Neel configuration through, e.g., Neel caps at the surface in nanoelements[116–119] or forming hybrid chiral Neel-Bloch walls in ferrimagnets.[120] Even the switching of a nominally 1D nanowire is mediated by transient 3D spin textures, i.e., Bloch points and Bloch lines, that have been predicted by micromagnetic simulations and are currently being explored experimentally.[121,122]

Nearly, all topological spin textures emergent in thin films, e.g., magnetic vortices and skyrmions, and their creation and dynamics have been so far interpreted within a 2D geometry model. Nevertheless, it has become evident that those textures extend across the depth of the film in a nonuniform manner, giving rise to features, such as twisted skyrmion tubes, magnetic boppers, or zippers.[123–129]

Whereas the topological charge is not affected by twisting skyrmion tubes, the topological Hall effect strongly depends on the twist angle effectively lowering its net contribution.[130] In addition to those isotropic solitons, systems with an inhomogeneous vector spin exchange[131] or an alternating Heisenberg exchange interaction between nearest and next-nearest neighboring spins are predicted to host anisotropic magnetic solitons[132,133] beyond dipole stabilized biskyrmions.[134,135]

Thus far, the vast majority of theoretical studies has been conducted by sole analytics, first-principle calculations or micromagnetic simulations with the primary goal to improve the understanding of nucleation and annihilation processes[136,137] and to find new ways to manipulate topological states in clean and ordered systems. Aside from magnetic field and current excitations, voltage control of anisotropy,[138,139] exchange, and strain[140] has emerged as a promising mean with first experimental demonstrations under way.[141–143] The challenge with modeling real materials is the existence of imperfections, defects, inhomogeneities, and structural/chemical disorder that require advances in all three avenues of theoretical studies. One recent example is the atomistic simulation of skyrmions in amorphous materials with interfacial DMI.[144]

Launching frustration into the third dimension allows for balancing the interaction between nearest and next-nearest neighbors of 2D artificial spin ice (ASI) systems, designing 3D geometrically frustrated systems, and envisioning novel types of spin exchange frustrated 3D spin textures, e.g., hopfions.[145–148]

In addition to the fundamental interest in stabilizing and manipulating a truly 3D topological knot, hopfions, which are in first approximation a twisted skyrmion tube whose ends join to form a tube, are technologically appealing since their current-driven lacks the gyrovector present in skyrmion dynamics that leads to a deflection toward the edges in a racetrack geometry.[149] This implies the absence of a topological Hall effect and calls for new detection mechanisms. Although hopfions are at the moment primarily a topic for theoretical models, first experimental approaches toward synthesis of hopfions in condensed matter materials and characterized using state-of-the-art instrumentation are emerging.

Furthermore, beyond investigating the inherent 3D spin textures in systems with nominally 2D and 1D shapes, advanced fabrication tools start empowering us to sculpt magnetic materials in full 3D with nanometer precision, opening the path to creating three dimensional surfaces and geometries that exploit shape to control spin textures in a whole new way.

Finally, numerous theoretical efforts enabling the modeling and predictions along these directions are emerging. They incorporate effects such as curvature-induced magnetochirality,[67,150] where the breakdown of inversion symmetry associated with the geometrical curvature leads to the emergence of a DMI-like spin interaction. Such an effect, just starting to be explored experimentally,[151] could lead to the stabilization of skyrmions and other complex spin textures,[152–155] thus providing a platform for magnonic crystals[156] or enabling curvature-induced domain-wall motion.[157]

II. PERSPECTIVE: SYNTHESIS OF 3D MAGNETIC SYSTEMS

Whereas the synthesis of 0D, 1D, and 2D nanomagnetic systems has become a mature technology, e.g., via lithography or thin film deposition techniques, e.g., molecular beam epitaxy (MBE), stepping beyond 2D magnetic systems in a tailored manner poses major challenges, particularly in terms of nanofabrication, due to the lack of standard tools for shaping and interfacing materials in 3D.

Looking into the near term future, there seems to be two main branches of current developments: (i) scalable platforms, compatible with the production of high-density economically viable devices at the cost of geometrical simplicity and (ii) rapid-prototyping techniques, which can be used to quickly explore complex 3D geometries for fundamental studies at the cost of not being scalable for mass production. Of course, the distinction of these two branches of development is merely a result of the view of the authors and does not signify, for example, that scalable techniques are not being used for fundamental studies. The classification of techniques as scalable or not is also a result of taking a snapshot of the field as of today; due to rapid technological developments, this classification may change unexpectedly.

A. Scalable and self-assembly techniques

Compact arrays of long and straight cylindrical nanowires present one of the most promising platforms to densely store magnetic information. As recently reviewed in detail,[24] several techniques exist for the fabrication of cylindrical nanowires with high-quality magnetic materials, including the possibility of producing core-shell structures as well as longitudinal variations in nanowire diameter and composition [Fig. 2(a)].[158] Recent results of current-induced motion of the Bloch-Point domain in $Co_{30}Ni_{70}$ nanowires grown in anodized aluminum oxide templates,[159] the realization of a magnetic ratchet[160] and the fabrication of interconnected networks[161] are examples of the increasing maturity of these techniques [Fig. 2(c)].

Since the early days of thin film growth, strain associated with different lattice constants, growth rates, and expansion coefficients had been seen as an inevitable byproduct. However, strain engineering has become a popular approach to manufacture 3D geometries out of virtually any material with virtually any shape [Fig. 2(b)]. This versatility enables to fabricate metamaterials, capacitors, robotics, electronics, terahertz transmitters, and magnetic sensors as highlighted in recent review articles.[162–165] Compared with aluminum template anodization, strain engineering takes advantage of conventional synthesis, lithography, and nanofabrication techniques and provides straight-forward integrability with minimal structural, electronic, and magnetic imperfections at the expense of typically larger dimensions, e.g., micrometers. While these dimensions are yet too large to cause theoretically predicted curvature effects, the 3D geometry still alters magnetic properties. A prominent example is the formation of azimuthal magnetization configurations in microtubular architectures with minimal magnetic anisotropy that boosts the giant magneto impedance effect (AC-variant of giant magnetoresistance) by several of orders of magnitude compared with planar stripes.[166]

Self-assembly of colloids in solution, e.g., Janus particles and superparamagnetic nanoparticles, is a fast yet versatile approach to

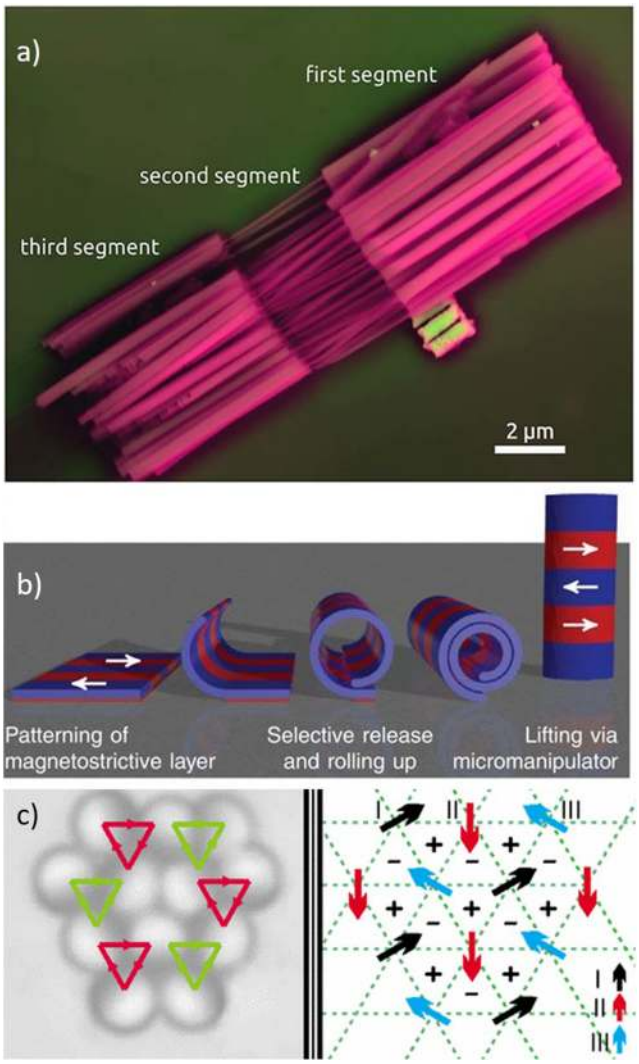

FIG. 2. Synthesis approaches for 3D nanomagnetic systems. (a) Scanning electron micrograph of a bundle of modulated NiCo wires isolated from their template (from Ref. 158). (b) Fabrication of azimuthally magnetized tubular architectures via selective rolling up of a prepatterned strained nanomembrane (from Ref. 115). (c) Image of a magic 12-particle cluster consisting of long-range-interacting colloidal particles with predefined magnetic moments (left) and schematics of the arrangement of the magnetic moments (right) (from Ref. 144).

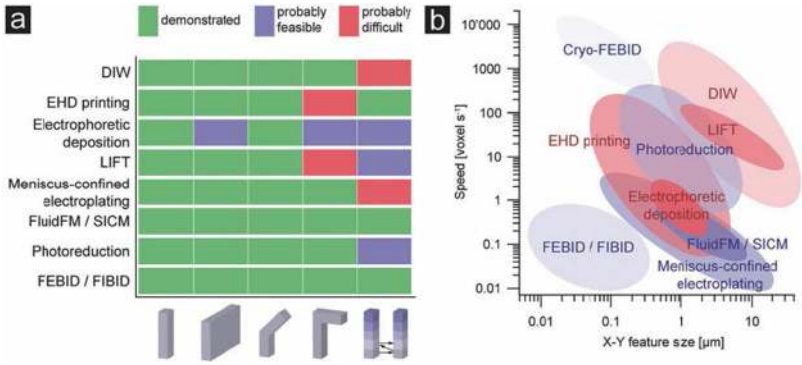

FIG. 3. Overview of capabilities and characteristics of micrometal additive manufacturing techniques (from Ref. 173). (a) Range of geometries enabled by each technique. (b) Speed vs x-y feature size of all techniques. The speed has been normalized by the voxel size.

prepare three-dimensional lithographic masks[167,168] and to study dipolar spin systems on the microscale and macroscale. The latter includes colloidal magnetic crystals with glassy properties[169] static[170] and dynamic self-assembly[167] within an AC magnetic field, and around a solid cylinder.[171] These examples reflect the current primarily fundamental aspect of self-assembled colloids. Recent research in 3D printing of liquids that possess magnetic properties of a solid magnet may open up a new path toward reconfigurable and reprogrammable magnetic devices.[172] Their novel functionalities rely on a reversible paramagnetic-to-ferromagnetic transformation of ferrofluid droplets by the jamming of a monolayer of superparamagnetic nanoparticles assembled at, e.g., water-oil interfaces. Both shape and magnetic moment have been shown to be reconfigurable. It remains to be seen whether the current micrometer sized droplets can be scaled down to and controlled at fundamental and application-relevant length scales for spintronics which would be in the tens of the nanometer regime.

B. Rapid prototyping of complex geometries

Being able to rapidly iterate over different complex geometries is of great value for the development of functional devices and the discovery of novel physical phenomena. The first way in which this is being pursued is via direct 3D nanofabrication of metals. A wide range of techniques is currently under development for that purpose (including magnetic materials) with submicrometer resolution.[173] The geometries enabled by the different tools are highlighted in Fig. 3(a), and resolution vs writing speed is illustrated in Fig. 3(b). Amongst these tools, Focused Electron Beam Induced Deposition (FEBID)[29] currently provides the best equilibrium in terms of resolution and availability of ferromagnetic materials, with a wide range of magnetic applications demonstrated,[88] including the recent demonstration of 3D building blocks for spin-ice systems.[174] One of the main limitations in FEBID nanofabrication of 3D nanostructures is the purity and microstructure of the deposited materials. Purities up to 90% have been demonstrated using FEBID deposition of cobalt;[175,176] however, such high purities are not generally the case for other materials and additional purification steps such as thermal annealing[177–179] are under development to improve material properties.

An emerging workaround to limitations in material availability is the combination of 3D patterning techniques with well-established material processing techniques such as electrodeposition[180] (Fig. 4) or thermal evaporation.[181] Albeit partially constraining the accessible geometries due to shadowing during thermal evaporation or the need for electrodeposited materials to enter the scaffolds, these hybrid nanoprototyping techniques have recently enabled the first demonstration of controlled domain-wall transport from a 2D substrate into a 3D nanostructure[181] (Fig. 5) thanks to the high-quality of the available materials and have also unlocked the creation of large-scale 3D frustrated nanowire lattices[182] of great interest for artificial spin-ice systems.

Complementary post processing techniques are under development for these systems in this case to reduce the accessible feature

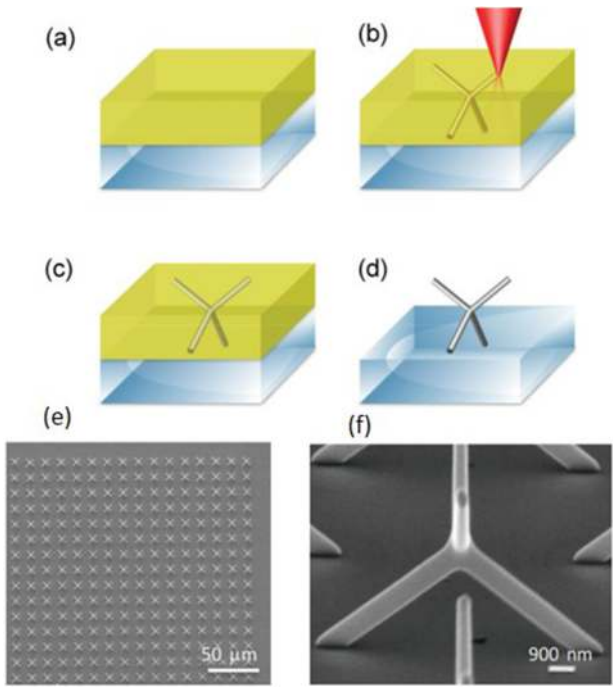

FIG. 4. (From Ref. 180) Two-photon lithography and electrodeposition. (a) Spin-coating of a positive resist onto a glass/ITO substrate. (b) Two-photon lithography of a 3D structure into the positive resist. (c) Electrodeposition of Co into the channels. (d) Lift off of the resist. (e) Large scale SEM of a tetrapod array (top view). (f) SEM image of a tetrapod obtained after 45° out-of-plane substrate rotation.

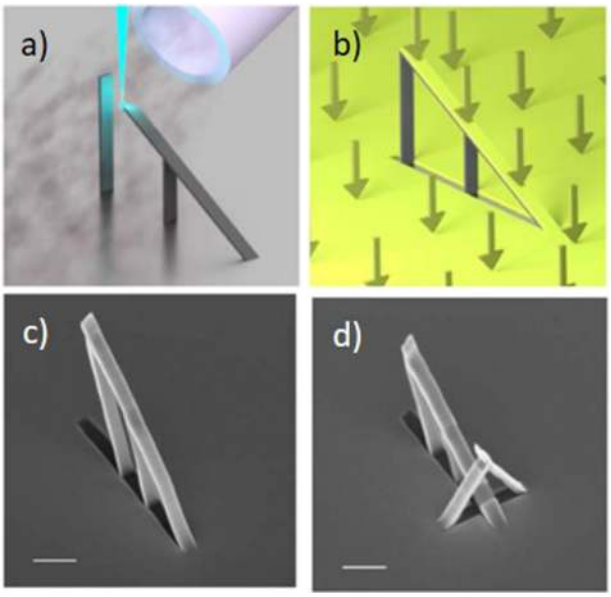

FIG. 5. (From Ref. 181) Fabrication of a 3D magnetic domain wall conduit. (a) 3D printing of a nonmagnetic scaffold using focused electron beam induced deposition. (b) 3D magnetic nanowire created by depositing a magnetic layer using thermal evaporation. (c) SEM image of a nanomagnetic conduit after Permalloy evaporation (50 nm). (d) SEM image of a control nanostructure, disconnected from the 2D source by adding a nanobridge at its base, which shadows the growth of evaporated magnetic material on the area beneath it. Scale bars 1 μm.

sizes. In a recent example, isotropic plasma etching and pyrolysis have been employed to reduce feature sizes of two-photon lithography scaffolds.[183] Recent advances in implosion nanofabrication[184] are another route that opens exciting paths toward the creation of complex 3D magnetic nanostructures.

III. PERSPECTIVE: CHARACTERIZATION OF 3D MAGNETIC SYSTEMS

The magnetic characterization of 3D nanoscale structures with atomic spatial resolution poses a challenge of similar magnitude as their fabrication due to uneven thickness and surface inhomogeneities inherent to 3D objects. In addition, when increasing shape complexity, the corresponding shape anisotropy acquires a nontrivial 3D structure which can favor complex spin configurations; this contrasts to 2D systems, where shape anisotropy favors the alignment of spins within a single plane.

Regarding these characterization challenges, we identify two complementary routes of development: (i) high-resolution techniques in which spins (and their direction) are directly imaged within a material using complex instruments. The most powerful tools are advanced X-ray spectromicroscopies taking advantage of the increased coherence of future x-ray sources, such as X-ray ptychography and holography at diffraction limited storage rings and X-ray free electron laser, or high-harmonic generation based tabletop x-ray microscopies and advanced aberration corrected transmission electron microscopies, 4D Scanning Transmission Electron Microscopies (STEMs) and electron holography approaches. (ii) Advanced benchtop magnetometry techniques rely on advanced laser-based, electrical or near-field probes to indirectly infer the magnetic configuration of nanostructures. High-resolution techniques are essential to precisely characterize the complex behaviors on the nano/atomic scale in 3D, i.e., correlate magnetic with structural, chemical, and electronic properties. The ultimate characterization would also involve studies of the spin dynamics down to ultrashort time scales in the fs regime with 3D spatial resolution. These high-end tools will be pushing the boundaries in addressing fundamental questions related to 3D nanomagnetism, but for technological applications and scale up processes, they would be rather impractical for every-day sample characterization. Toward that end, benchtop instruments that, sacrificing on resolution or relying only on stray fields, can be employed to infer the magnetic alignment of spins within complex 3D samples are under development. Both are discussed in the following.

A. Tomographic and high-resolution direct magnetic characterization

X-ray vector field tomography has seen a tremendous development in recent years to visualize the magnetization vector field in 3D with both hard and soft x-ray sources (Fig. 6).[185–188] These efforts were complemented using conventional shadow contrast x-ray photoemission electron microscopy (X-PEEM) to image the magnetization configuration in 1D nanorods.[122,189] Electron techniques, such as holography and Lorentz microscopy, allowed for mapping the magnetic states in nanocubes[190,191] and nanowires[192] and visualizing anisotropic skyrmion phases in extended thin films.[193,194]

There are three different routes to advance magnetic imaging capabilities: (i) enhance spatial resolution and contrast/sensitivity with phase contrast imaging, (ii) improve performance of tomographic imaging (experimental and computational), and (iii) enable time-resolved pump-free imaging at the microsecond time scale. Either thrust faces challenges with sensing a magnetic contrast that is relatively weak compared with electrostatic/structural contributions or will strongly benefit from ongoing developments of faster, more sensitive detectors, e.g., direct electron detectors, and better sources, e.g., monochromatic, brilliant, and coherent beams, offered by aberration-corrected transmission electron microscopes and next-generation diffraction-limited light sources. Coherent electron and x-ray beams are particularly crucial to phase contrast imaging based on exit wave reconstruction[120] and coherent diffraction imaging techniques, such as ptychography.[195,196] To date, a coherent x-ray beam is generated with an aperture that clips more than 90% of beam thereby significantly reducing the temporal resolution of coherent diffraction imaging and x-ray photon correlation spectroscopy. Better probes and faster detection will further require more efficient software algorithms and data handling/analysis, including, in particular, live updates based on automated sample alignment, data collection, and reconstruction. This will not only allow for studying magnetic systems in detail but also improve the accessibility to a limited number of high-end instruments.

B. Benchtop magnetometry and indirect characterization techniques

Within benchtop characterization techniques (Fig. 7), magneto-optical Kerr effect (MOKE) magnetometry and magnetic force

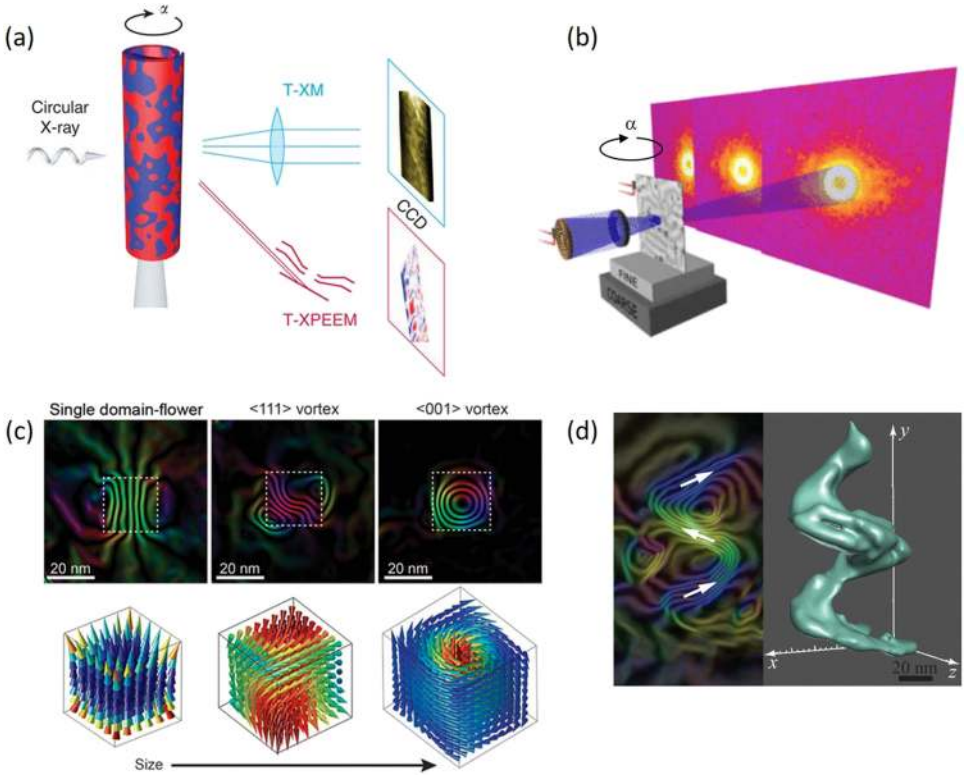

FIG. 6. 3D tomographic characterization techniques with high spatial resolution is extensions of advanced x-ray and electron microscopies. (a) X-ray tomography in real space uses full-field transmission x-ray microscopy and transmission (shadow) X-ray photoemission electron microscopy (X-PEEM) techniques (from Ref. 115). (b) Reciprocal space x-ray imaging can extend x-ray ptychography toward 3D tomography both for soft and hard x-rays (from Ref. 188). (c) Electron holography images of size-dependent magnetization configurations in single-crystalline Fe nanocubes (from Ref. 191). (d) Electron holography image of the magnetization distribution inside a sculpted three-dimensional Co nanospiral (from Ref. 192).

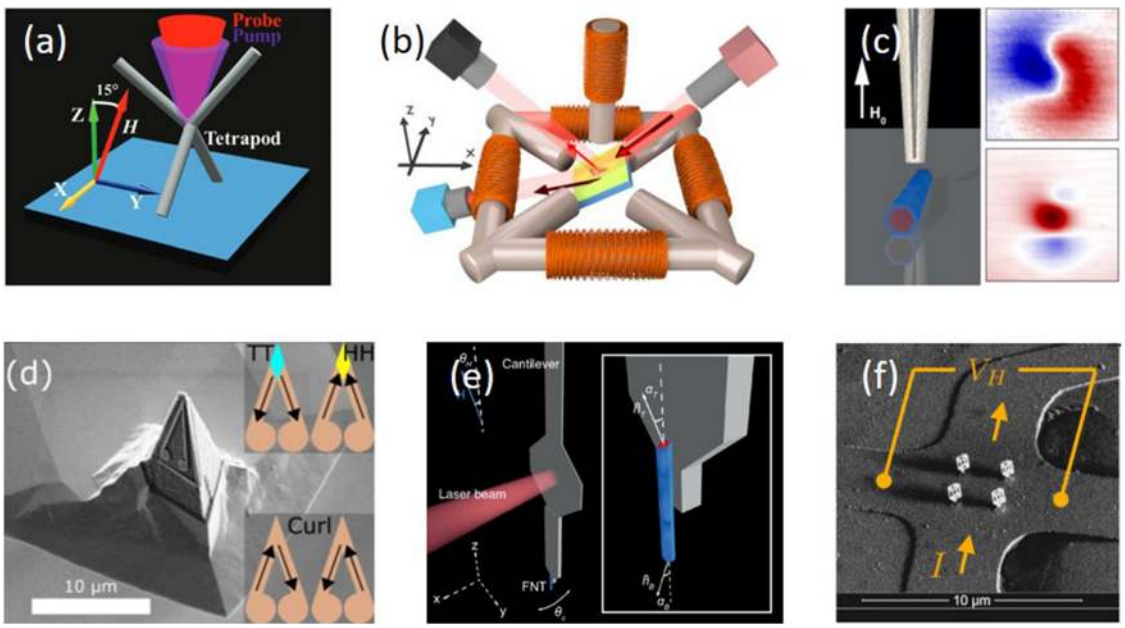

FIG. 7. Various magnetometry and scanning probe benchtop characterization tools have been developed for the characterization of 3D magnetic nanostructures. (a) Time resolved MOKE to probe spin-wave modes of a junction in a single tetrapod structure (from Ref. 202). (b) Schematics of dark-field MOKE (from Ref. 181). (c) Imaging stray magnetic fields of individual magnetic nanotubes with a scanning nano-SQUID (from Ref. 198). (d) Reconfigurable FIB-patterned MFM tip to study domain walls (from Ref. 200). (e) Observation of end-vortex nucleation in single magnetic nanotubes with a dynamic cantilever (from Ref. 203). (f) SEM micrograph of a micro-Hall cross and measurement configuration (from Ref. 204).

microscopy (MFM) have historically been two of the most widely used techniques in 2D magnetism. Albeit generally limited to their top surfaces, MFM of 3D magnetic nanostructures is now commonplace, with the development of many custom probes that enhance resolution and reduce tip-sample obstruction under way.[197–200] Integration advances such as the development of AFM setups that can be incorporated inside scanning electron microscopes for precise tip placement[201] may potentially remove one of the greatest constraints in MFM characterization of 3D nanostructures. MOKE magnetometry has also been recently employed to probe the top surfaces of a 3D nanowire lattice[182] and the dynamics of a single tetrapod structure.[202] Standard MOKE magnetometry, however, suffers from the fact that measured signals are integrated over the relatively large size of the laser spot employed (typically in the micrometers rage). In a paradigm change for magneto-optical characterization of 3D nanostructures, dark-field MOKE magnetometry has been recently developed.[181] In this technique, the 3D nature of the sample is exploited to gather the different reflections created by different surfaces in the sample. This allows us to characterize in parallel different parts of a nanostructure that have close to no spatial separation between them, using a probe much larger that the structure itself.

Less-common probes, such as scanning nano-SQUIDs[198] and dynamic cantilever magnetometers,[203] have also been employed recently to characterize 3D nanomagnetic structures, and micro-Hall effect setups have also been demonstrated.[174,204]

IV. CONCLUSIONS

The science of nanoscale magnetic materials, which over the last few decades has not only contributed to a deeper and fundamental understanding of magnetism but was the key enabler for numerous technologies that dominate the life and business in today's society, is on the verge of entering another dimension with tremendous transformative potential. Building on the accomplishments from nanomagnetism in reduced dimensions, i.e., harnessing proximity and confinement, there is now the opportunity to use those in the third spatial dimension. Although this will open a path for more complexity, functionality, and novel behavior of nanomagnetic materials and their technological applications, it will require novel approaches both in synthesis, modeling, and theory and particularly in the validation, i.e., for characterization of those three-dimensional magnetic systems.

It can be expected that the combinatorial manifold, e.g., in multicomponent materials in three dimensions will go beyond a linear expansion of the current material design. The ability to tailor the couplings and interactions of neighboring spins in 3D should lead to very interesting and novel spin textures, including higher levels of frustration, e.g., in 3D spin ice systems, that will have no counterpart in conventional bulk systems. Topological effects, which recently have stimulated intense research activities into magnetic skyrmions for both fundamental and applied reasons, will likewise expand into highly complex topological spin textures, such as hopfions.

The dynamics of those three-dimensional nanoscale spin systems, specifically fluctuations across time scales with short spatial correlations, will be another area, where the tailored synthesis and modeling will open new avenues for research.

Ultimately, embracing the capabilities in 3D nanomagnetism will very likely trigger the development and emergence of novel applications, that will take full advantage of all three spatial dimensions, and will not just save footprint in future technologies.

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