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2025 Roadmap on 3D Nano-magnetism

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Abstract

The transition from planar to three-dimensional (3D) magnetic nanostructures represents a significant advancement in both fundamental research and practical applications, offering vast potential for next-generation technologies like ultrahigh-density storage, memory, logic, and neuromorphic computing. Despite being a relatively new field, the emergence of 3D nanomagnetism presents numerous opportunities for innovation, prompting the creation of a comprehensive roadmap by leading international researchers. This roadmap aims to facilitate collaboration and interdisciplinary dialogue to address challenges in materials science, physics, engineering, and computing.

The roadmap comprises eighteen sections, roughly divided into three parts. The first section explores the fundamentals of 3D nanomagnetism, focusing on recent trends in fabrication techniques and imaging methods crucial for understanding complex spin textures, curved surfaces, and small-scale interactions. Techniques such as two-photon lithography and focused electron beam-induced deposition enable the creation of intricate 3D architectures, while advanced imaging methods like electron holography and Lorentz electron Ptychography provide sub-nanometer resolution for studying magnetization dynamics in three dimensions. Various 3D magnetic systems, including coupled multilayer systems, artificial spin-ice, magneto-plasmonic systems, topological spin textures, and molecular magnets are discussed.

The second section introduces analytical and numerical methods for investigating 3D nanomagnetic structures and curvilinear systems, highlighting geometrically curved architectures, interconnected nanowire systems, and other complex geometries. Finite element methods are emphasized for capturing complex geometries, along with direct frequency domain solutions for addressing magnonic problems.

The final section focuses on 3D magnonic crystals and networks, exploring their fundamental properties and potential applications in magnonic circuits, memory, and spintronics. Computational approaches using 3D nanomagnetic systems and complex topological textures in 3D spintronics are highlighted for their potential to enable faster and more energy-efficient computing.

Keywords: Nanomagnetism; three-dimensional nano structures, fabrication techniques, imaging methods; analytical methods, computational approaches.

Introduction

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In the burgeoning field of nanoscience and nanotechnology, the manipulation of matter at the nanoscale has paved the way for extraordinary advances in multiple fields. One particularly fascinating area that has attracted huge attention is the three-dimensional (3D) nanomagnetism. Nanomagnetism deals with the study and manipulation of magnetic phenomena at lateral dimensions of nearly 100 nanometers or less. When the size of magnetic structures shrinks to the nanometer scale, their magnetic properties change drastically. In the 3D range, the complexity and richness of magnetic interactions increase significantly due to the intricate spatial arrangement at the nanoscale, offering a wealth of opportunities for both fundamental research and practical applications. The interplay of shape, size, and material composition in 3D configurations leads to phenomena such as complex magnetization reversal modes, topology and frustration, novel magnetic textures as well as magnetization dynamics.

As traditional two-dimensional (2D) approaches face limitations, the exploration of 3D magnetic architectures promises increased data storage densities, non-volatility and scalability, enhanced energy efficiency, and the potential for revolutionary computing paradigms. Spintronics, sensors, and quantum information processing are just among the large range of areas where the principles of 3D nanomagnetism are reshaping technological landscapes. Fig. 1.1 shows a series of free standing 3D magnetic nanostructures, complex spin textures, interconnected conduit networks and 3D nanomagnetic circuits based on a nanobridge geometry.

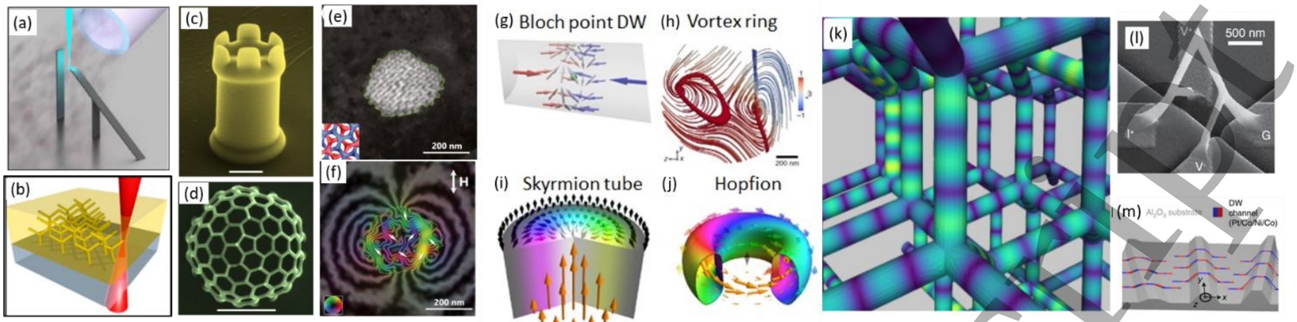

Figure 1.1 (a) Focussed electron beam induced deposition (FEBID) [1] and (b) Two-photon lithography method [2]. By the controlled beam movement, bulky (c) and complex buckyball like (d) structures can be realized using FEBID [3,4]. Phase contrast imaging with off-axis and in-line electron holography using aberration-corrected transmission electron microscopy. (e) Structure and (f) magnetic induction of 3D gyroid network visualized with off-axis holography [5]. Schematic spin textures of (g) a Bloch point type domain wall [6], (h) vortex ring [7], (i) a skyrmion tube, and (j) a Hopfion with *Hopf index* = 1 [8]. (k) Various oscillation modes in a diamond-type artificial magnetic crystal developing at different frequencies [9]. Exemplary 3D spintronics devices showing (l) ferromagnetic interconnector directly printed by FEBID [10] and (m) state-of-the-art racetrack memories formed by synthetic antiferromagnets, positioned on 3D pre-patterned substrates to achieve vertical domain wall motion [11].

Furthermore, the roadmap of 3D nanomagnetism recognizes the pivotal role of novel characterization tools with high temporal and spatial resolution, alongside sophisticated computational modeling in driving progress. The synergy among theoretical modeling, synthesis and fabrication, as well as characterization and validation opens a path towards exploitation of 3D nanomagnetism in future technological applications. Through collaborative efforts and strategic initiatives, our goal is to unlock novel frontiers in science, engineering, and technology heralding a paradigm shift in comprehending and harnessing magnetic materials in three dimensions.

This Roadmap sets the stage for an in-depth exploration of the fascinating world of 3D nanomagnetism and aims to provide a clear and concise overview and reference points to researchers from academia, research institutes and industry, and funding agencies a comprehensive understanding of 3D nanomagnetism. It provides guidance on foundational knowledge, state-of-the-art techniques, and emerging trends, helping to cultivate a skilled workforce capable of driving future advancements in the field. By understanding the intricacies of magnetic behaviors in 3D, researchers and engineers are poised to unearth new frontiers in technology, heralding a future where nanoscale magnetic structures play a crucial role in shaping the next generation of electronic and information processing systems.

This 2024 Roadmap on 3D nanomagnetism is inspired by the rapidly emerging research activities in the field of magnetic nanostructures in recent years and follows other Roadmaps for magnetism, [12,13] magnonics, [14,15] review articles [16,17,18] and book.[19]

It comprises eighteen contributions produced by thirty nine leading international researchers involved in specific scientific and technological domains and is divided into three main blocks, namely (i) Fabrication and Characterization, (ii) Analytical and Numerical Methods and (iii) Applications of 3D Magnetic Nanostructures. Each block is divided into sections and each section describes the background of the topic, current and future challenges, and advances in science and technology to address these challenges. Finally, future research directions and possible strategies for each author's particular area of interest are presented.

The first block consists of 10 sections and deals with the fabrication and characterization of 3D magnetic nanostructures, covering recent advances in fabrication methods and imaging techniques emphasising the importance of understanding complex spin textures and curved surfaces as well as small-scale interactions.

In **Section 1**, *Ladak*, *Bran*, and *Grundler* review both top-down and bottom-up fabrication methods to realize 3D magnetic nanostructures on length scales of 100 nm and below and present future challenges that need to be addressed. They further discuss how optimizing the fabrication of 3D magnetic nanostructure is crucial for the discovery and understanding of complex topological spin textures, their interactions, and time-dependent responses.

In **Section 2**, *Huth* and *Plank* review of the unique capabilities of 3D- focused electron beam induced deposition (FEBID) as a reliable 3D nanoprinting technology. They also present different strategies for the 3D nanomagnet fabrication, and explain how 3D growth simulations are necessary for the systematic analysis of shape-controlling factors.

Section 3 by *Schmidt* and *van Dijken* provid an overview on the recent developments of 3D suspended yttrium iron garnet (YIG) structures which have already achieved a state beyond the mere proof of principle. Structures have been fabricated that theoretically fulfil the requirements for optical photon-magnon coupling and the first mechanical resonances have been measured despite relatively low frequencies.

Section 4 by *Streubel* and *Dobrovolskiy* explore the possible pathways for correlating the impact of 3D structure, i.e., shape, curvature, interfaces, and defects, on the electronic and magnetic properties of 3D nanomagnets. Special emphasis is placed on the requirements of advanced characterization techniques that combine high sensitivity with spatial and temporal resolution.

Scagnoli, *Heyderman*, and *Donnelly* present in **Section 5** an overview of 3D nanomagnetic imaging used to capture, reconstruct, and analyze the magnetic behavior of and topological features within materials and microstructures in 3D with nanoscale precision. Unlike traditional magnetic imaging techniques that operate at larger length scales (such as Kerr microscopy), or give information only from surfaces or average signal from thin films (such as Lorentz transmission electron microscopy), advanced 3D nanomagnetic imaging methods enable the vectorial visualization of magnetic domains and intricate spin textures such as vortex loops, skyrmions and Bloch points governing the behavior of magnetic nanomaterials.

Section 6 by *Hellwig* and *Fallarino* review how magnetically coupled multilayer systems allow to access and to explore new energy scenarios and, consequently, how novel degrees of freedom for a widespread variety of applications can be achieved by varying and designing innovative material properties. From this prospect, the first targeted efforts towards the stabilization of complex 3D individual magnetic textures as well as interconnected periodic 3D architectures are just emerging. In addition, a brief overview on the interest toward stacking of layered 2D magnets is presented.

In **Section 7**, *Jungfleisch* and *Farhan* introduce currently popular activities in the field of artificial spin ice (ASI) systems and how the formation of 3D ASI is increasingly gaining momentum. To this end, potential future endeavors are introduced to allow for 3D ASI systems to undergo full-scale characterization, both for researchers interested in statistical physics and magnetization dynamics.

Maccaferri and *Vavassori* discuss, in **Section 8**, how the combination of the uniqueness of the 3D photonic band structure with the peculiarities of 3D spin textures may open new avenues for much improved control over magneto-optical and opto-magnetic effects. The authors also present the challenges associated with the transition from 2D to 3D magneto plasmonics and the pathways envisioned to overcome them.

In **Section 9**, *Fischer, Tomasello*, and *Finocchio* focus on the emerging topic of 3D topological spin textures as a fascinating frontier in the study of condensed matter physics. These intricate patterns of spin configurations, governed by the principles of topology, exhibit robustness against local perturbations and hold promise for realizing fault-tolerant information storage and processing. By harnessing the unique properties of topological spin textures, researchers aim to develop innovative techniques for manipulating and controlling spin degrees of freedom in three dimensions, thereby enabling advanced functionalities such as topological spintronics and quantum information processing.

In **Section 10** *Sessoli* and *Clérac* address the molecules as a still underexplored resource for nanomagnetism. The chemical design can control several key features, ranging from magnetic anisotropy to exchange interaction, thus resulting in high coercivity. The intrinsic confinement at the nanoscale and the easy processability make magnetic molecules an ideal playground for the development of spin-based quantum technologies.

The second block comprises 3 Sections about analytical and numerical methods to describe the magnetism of 3D nanostructures and curvilinear systems.

In **Section 11,** *Makarov* and *Sheka* offer an overview of prospective advancements in the field of curvilinear magnetism. This progress is likely to be influenced by the active exploration of geometrically curved magnetic architectures in both fundamental research and technology.

Section 12 from *Krawczyk, Gallardo*, and *Landeros* present an overview of the analytical methods used to describe 3D magnetic nanostructures, discussing their scope, limitations, and future projections. The importance of the variation of magnetization in the volume, the central role of the magnetic dipole interaction, and the importance of the boundary conditions are emphasized.

Section 13 by *d'Aquino* and *Hertel* discuss the challenges in recent numerical approaches to simulate the magnonic properties of artificially structured 3D nanomagnets, focusing on interconnected nanowire arrays and their high-frequency magnetization oscillation modes. This highlighted the need to use finite-element-based algorithms for such complex geometries with low volume occupancy and the advantages of solving magnonic problems directly in the frequency domain with appropriate large-scale methods instead of simulating magnetization dynamics in the time-domain by integrating the Landau-Lifshitz-Gilbert equation. Recent examples of simulation studies utilizing these techniques are also presented and discussed.

The final and third, block consists of 5 Sections.

Barman and *Gubbiotti* give an overview, in **Section 14**, of the recent progress and the future possibilities of 3D magnonic crystals which permit novel degree of freedom to control, manipulate and guide the spin-wave that are topographically impossible in 2D systems. Vertical stacking offers versatile coupling conditions, i.e., dipolar and interlayer exchange coupling due to the smallest possible spacing as well as access to non-reciprocity arising from the interfacial Dzyaloshinskii Moriya interaction and the topography of the curved magnonic conduits.

In **Section 15**, *Pirro* and *Ciubotaru* examine the potential benefits of magnonic circuits in terms of energy consumption and showed how the energy requirements could be reduced by one to two orders of magnitude by moving to a 3D configuration. The authors also presented the schematic of a potential 3D magnonic network consisting of 2D layers connected by 3D directional couplers and discussed the fundamental and technological advances that are still needed to turn 3D magnonic circuits from concept to reality.

Section 16 concerns with the computation in 3D nanomagnetic systems. Gartside and Becherer provide an overview of a large variety of magnetic computation schemes and highlight possible routes forward that harness uniquely available benefits by moving magnetic systems into the third dimension and avoiding the more generic advantages of 3D, such as increased density.

Section 17 by *Ono* discusses how 3D magnetic memory a significant advancement in data storage technology, promises higher storage densities and improved efficiency compared to traditional planar memory. By utilizing all three dimensions of space for data storage, this technology allows for packing more information into a smaller physical footprint. As the digital world continues to generate vast amounts of data, 3D magnetic memory stands out as a promising solution to meet the escalating storage needs of today and the future.

Finally, **Section 18** by *Fernández-Pacheco* and *Bortolotti* focus on the topic of 3D spintronics. By extending spin-based operations into three dimensions, this technology opens avenues for compact, high-performance devices with enhanced processing power and storage capacity. Through intricate manipulation of spin states in nanostructures, including complex 3D topological textures, three-dimensional spintronics promises faster and more energy-efficient computing, paving the way for transformative innovations that could redefine the landscape of modern technology.

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1- Nanofabrication of 3D Nanomagnets

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1.1. Status

In 2008, Stuart Parkin *et al* envisaged a new data storage device, named magnetic racetrack memory, which harnessed the injection and propagation of domain walls in a ferromagnetic 3D nanowire network, in order to read and write information [1]. At the time of conceptualisation, the field of 3D nanomagnetism had already witnessed the creation of ferromagnetic nanocrystal superlattices, vertically standing nanowires and individual nanotubes, but the proposed 3D device architecture was visionary.

Recently, a number of distinct research strands have yielded a new paradigm in nanomagnetism whereby mastering of the third-dimension was advanced [2]. The paradigm resulted in new physics and applications, providing novel spin textures stabilized by novel geometry, topology and frustration as well as their time-dependent phenomena. This has partially been driven by the development of new theories, which suggested new terms in the free energy of a ferromagnet due to local curvature and torsion [3]. In addition, both top-down and bottom-up fabrication methods have matured allowing realization of 3D structures on length scales of 100 nm and below. In the following, we discuss state-of-the-art fabrication methods which are split into those associated with direct-write technologies and those associated with self-assembly.

In focussed electron beam induced deposition (FEBID), a gaseous precursor containing metallic content is injected into a scanning electron microscope [4]. Interaction between the electrons and the precursor yields a decomposition of the volatile components, and a local deposition of the metallic content [4]. By scanning the electron-beam with respect to the substrate, 3D metallic nanostructures can be directly written (Fig 1a-d). Another direct-write technology that uses photons instead of electrons is two-photon lithography (TPL). In this technique, a femtosecond pulsed laser, is focussed to a diffraction-limited spot within a photo-resist, as illustrated in Fig 1e [5]. Polymerisation of the resist only occurs within the central portion of the focal volume. One scans the beam in order to realize arbitrary structures within the resist. The polymeric structures as well as porous templates have then been combined with line-of-sight deposition to realise 3D artificial spin ice (Fig 1f), atomic layer deposition [6] to realise 3D nanomagnetic networks for magnonics (Fig 1g) or electrodeposition. Another approach is to harness strain engineering to self-assemble non-trivial ferromagnetic geometries such as rolled up tubular structures [3,7] as shown in Fig 1(h,i). Such structures exploit curvature to realise novel spin textures. Finally, combining anodised alumina or ion-track etched templates with electrochemical deposition allows the realisation of cylindrical or tubular magnetic nanowires [8,9] with both uniform (Fig 1j) and modulated geometry (Fig 1k). Overall, recent developments in nanofabrication technologies have enabled a wide range of intricate 3D nanostructures with unique magnetic properties and spin textures.

1.2 Current and Future Challenges

The advancement of 3D nanotechnology relies on the synthesis and fabrication of high-quality nanostructures with well-defined shapes, high-purity materials, and good interfacial properties. Although significant progress has been made in recent years in the 3D fabrication process, there are still important challenges to overcome.

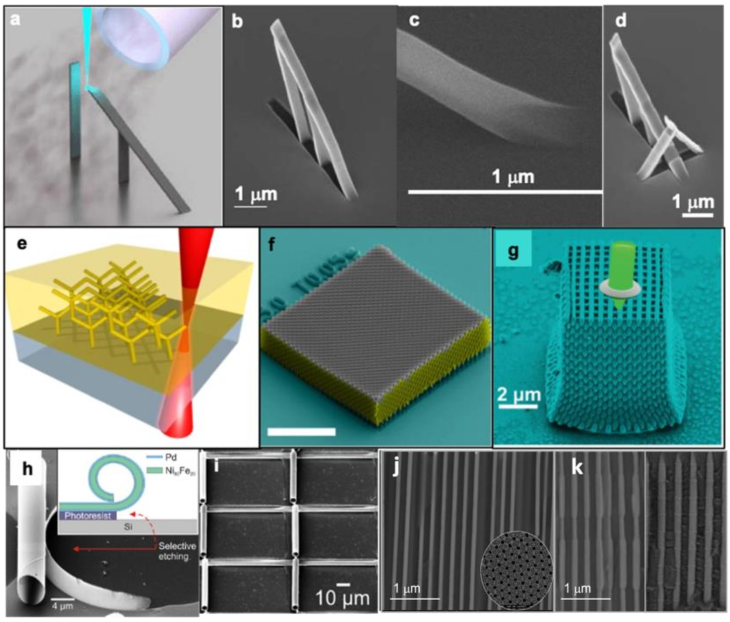

Figure 1.1. Overview of fabrication methodologies used to realise 3D magnetic nanostructures. (a) Focussed electron beam induced deposition, which has been used to realise (b-d) tilted 3D magnetic nanowires for domain wall injection [4] (e) Two-photon lithography which has been used to realise a range of structures including (f) a 3D artificial spin-ice [5] and (g) a 3D nanomagnetic network [6]. (h) Rolled-up ferromagnetic microtubes [7]. (i) Microtubes in a patterned array [7]. (j)-(k) FeCo nanowires with uniform and modulated diameters fabricated by electrodeposition into alumina templates (inset) [8].

Direct-write techniques such as FEBID and TPL, offer versatility with respect to geometry of structures, but both have challenges in order to yield high purity multi-material structures with well-defined interfaces. With FEBID, the key challenges of this technique are the reduced purity of the material as contaminants from the precursor molecules are embedded in the final structure and the limited number of available precursors for magnetic materials. TPL can be combined with electrodeposition to realise a wide-range of high-purity magnetic materials. In addition, use of high-speed galvo-mirrors allows the relevant polymeric templates to be fabricated at speeds of order mm/s. However, reaching sizes of order 10 nm that can be obtained with FEBID is very challenging due to diffraction.

Atomic layer deposition or chemical methods like electrodeposition combined with for instance anodised alumina or ion-track etched templates are reliable and allow for large arrays of high aspect ratio nanostructures like nanowires, nanotubes, core-shell and multi-layered nanowires with diameters between 20-300 nm formed by a large variety of materials with well-defined crystallographic structures. The main problem with the chemical methods is geometric limitations imposed by the templates, meaning it is difficult to realise 3D nanostructures of complex geometry.

All of the methods described above have considerable challenges to overcome in order to realise complex heterostructures with ultra-thin film thicknesses and smooth interfaces to achieve DMI or RKKY interlayer exchange coupling, as has been explored in planar systems. Today's planar technologies for magnetic memory and sensing involve furthermore interfaces with antiferromagnets and insulators forming ultrathin tunnel barriers, soft-magnetic components for guiding stray fields and hard-magnetic nanograins with specifically designed crystallographic orientation for non-volatile storage. Future challenges concerning functional 3D nanomagnetism revolve around finding a series of precursors and a combination of fabrication processes that are compatible with semiconductor processes and allow the intricate physics offered by 3D nanostructures to be exploited in devices for e.g. memory or sensing applications. For technological relevance, on the one hand, large-scale mass production strategies need to be explored. On the other hand, the integration with conventional electronics will be important. It requires the exploration of contact strategies in order to provide complex 3D devices with spin-polarised currents and / or time-varying magnetic fields to allow embedded functionality.

1.3 Advances in Science and Technology to Meet Challenges

In many cases, the fundamental science and technologies required to address the aforementioned challenges are already available but like many scientific challenges require extensive interdisciplinary research to find the correct implementation. Optimisation of FEBID deposition parameters can yield carbon contamination as little as 5%, whilst high temperature annealing increases the purity to almost 100%. With respect to diversifying FEBID precursors, the approach to date has been to utilise well-known systems already realised for thermal processes (e.g. chemical vapour deposition). A fresh approach is to design new precursors with electron-based decomposition in mind, and to characterise in detail electron-driven chemical changes using techniques such as x-ray photoelectron spectroscopy. Such studies have already produced bi-metallic precursors with high metallic content. TPL can harness a wide-range of techniques and approaches from non-linear optical microscopy in order to further improve feature size and speed. Utilization of shorter wavelength lasers, allows the realisation of sub-100 nm feature size, whilst exploiting spatial light modulators can allow the production of hundreds of foci, each writing structures in parallel. Replacing conventional anodized alumina or ion-track etched templates with those produced using TPL and harnessing chemical techniques are then a promising approach to realise 3D magnetic nanostructures of different architectures. Electrodeposition is already widely used in fabrication facilities to realise a range of pure magnetic and non-magnetic materials. A careful combination of process condition control during the TPL step and use of chemical cocktails, may allow the removal of the polymeric template. Alternatively, additional processes such as ALD allow for a conformal coating of individual nanowires and complex 3D nanoarchitectures to either protect these 3D nanomagnets from aggressive oxygen plasmas needed in subsequent fabrication processes or to add further free-form 3D magnetic elements [10].

1.4 Concluding Remarks

The optimization of 3D magnetic nanostructure fabrication is crucial to discover and then understand complex topological spin textures, their interactions and time-dependent response. Mastering the fabrication from the macroscopic down to the microscopic length scales will allow one to build networks of exotic 3D nanomagnets. We expect them to pave the way to the next generation of functional magnetic devices which allow for ultrahigh density non-volatile memory technologies or charge-less information processing by either pure spin currents or spin waves in 3D architectures.

Acknowledgements

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2 – Printing 3D Magnets at the Nanoscale

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2.1 Basics of 3D nanoprinting

With regard to design flexibility, achievable spatial resolution, hybride structure fabrication capabilities and – to a growing degree – materials selection, focused electron beam-induced deposition (FEBID) is one of the most capable methods for 3D nanomagnet fabrication. In FEBID, a gaseous precursor gas supplied via a dedicated gas injection system (GIS) is delivered to the focus area of a focused electron beam within a scanning electron microscope (SEM). Although all electrons have a certain probability to dissociate the adsorbed precursor molecules, low energy secondary electrons (SE) are mainly responsible for the intended growth by immobilizing the non-volatile fragments [**1**]. By careful control of the lateral electron beam movements and proper selection of dwell times, a wide range of even complex 3D shapes with sub-20 nm feature sizes can be fabricated in an additive single step procedure [**2**], as illustrated in Figure 1, further denoted as 3D Nanoprinting (**3DNP**). The material composition and microstructure are largely determined by the precursor-specific dissociation channels, as well as the patterning strategy [**3**]. With regard to 3D nanomagnet fabrication at least three pathways can be followed: (a) direct-write of magnetic 3D structures using a suitable precursor, see e.g. [**4**], (b1) fabrication of a 3D scaffold for subsequent physical vapor deposition (**PVD**) of a magnetic layer, see e.g. [**5**], or (b2) site-selective chemical vapor deposition (**CVD**), accomplished by passing a dissipative current through the scaffold that leads to local heating, allowing for thermally induced dissociation of appropriate precursor gas [**6**].

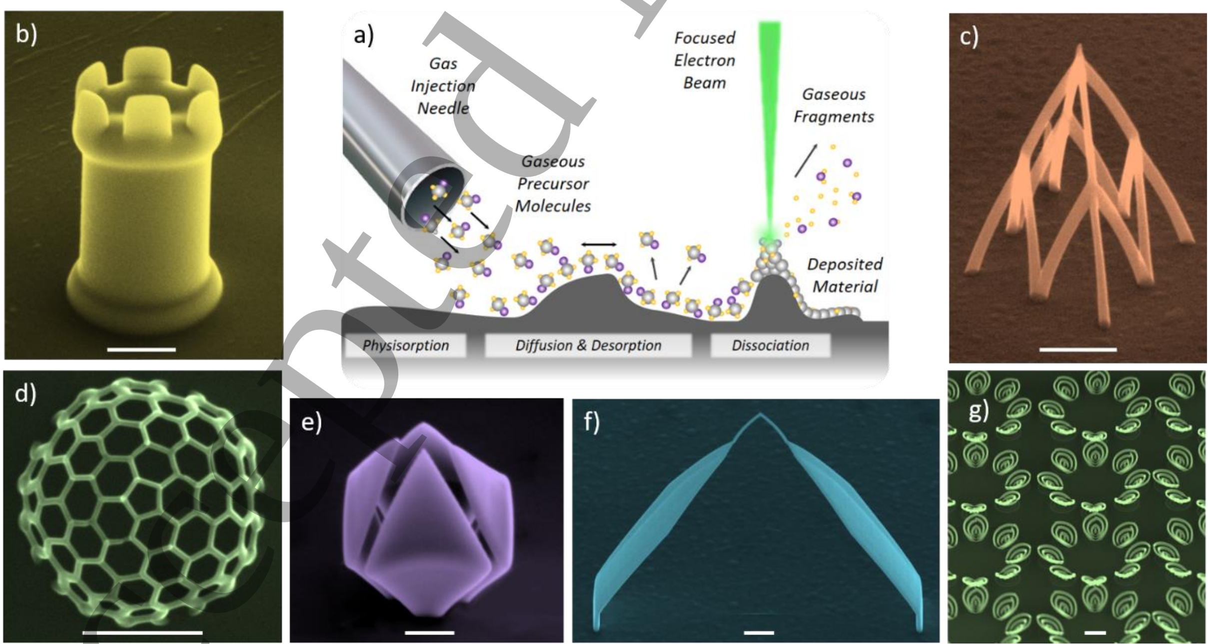

Figure 2.1. (a) Basic principle of FEBID, where a focused electron beam dissociates and immobilizes surface adsorbed precursor molecules. By the controlled beam movement, bulky (b) or mesh-like 3D structures (c) can be realized, allowing for more complex (d), sheet-like (e) and even combined (f) designs for fabrication of single objects (b-f) or larger arrays (g). All images are recolored, tilted SEM images with a 500 nm wide scale bar.

2.2 3D growth control

For a shape-true transfer of a 3D nanomagnet model to an actual 3D deposit several FEBID-specific aspects have to be considered. First, the electron-induced dissociation process is non-local, meaning that deposition will occur at any place where SEs reach the substrate or deposit surfaces, if precursor is available. Mostly, this is close to the primary beam impact region (desired, local deposit area), although SEs are also generated by backscatter electrons (BSE) and forward-scattered electrons (FSE). Second, the available precursor, i.e. adsorbate density, depends on nearby precursor consumption and diffusive replenishment, summarized in the term *working regime*. This leads to a complex interplay of growth-mode and deposit-shape dependent proximity effects, which can dynamically change with the growing 3D structure. Since the beam movement is laterally controlled, conceptual similarities to the approach used in other 3D printing methods do exist. In particular, slicing the 3D model perpendicular to the z-axes (growth direction) in order to extract the lateral beam positions for any given z-height is also used in 3D-FEBID (Figure 2). For any such slice, the discretization of the lateral beam positions (x_i, y_i) and associated local dwell times have to be chosen appropriately and are the key for high-fidelity manufacturing. For that, it may be necessary to compensate for the local precursor depletion by implementing a proximity correction, based on a 3D growth simulation in order to understand the dynamic coverage situation with implications on shape-determining factors, as explained below.

2.3 3D-FEBID growth simulation

For practical reasons, any 3D-FEBID growth simulation approach should be sufficiently fast to allow a systematic analysis of shape-controlling factors, such as local and non-local SE generation, temperature-gradient formation due to energy dissipation and spatially-resolved precursor density distributions as a consequence, to name a few. This rules out first principle simulation approaches, as well as those relying on molecular dynamics, as time scales involved in FEBID span from the femto-second (electron dynamics) to milli-second range (typical beam dwell times for 3D growth) and beyond. An approach that has been shown to fulfill the necessary criteria is numerically solving the diffusion-reaction equation for the precursor density n (normalized to the density of one monolayer n_0) on the surface of the growing deposit:

$$\frac{\partial n(x,y)}{\partial t} = k\Phi\left(1 - \frac{n}{n_0}\right) - \frac{n}{\tau} - \sigma f n + D\left(\frac{\partial^2 n}{\partial x^2} + \frac{\partial^2 n}{\partial y^2} + \frac{\partial^2 n}{\partial z^2}\right)$$

This has to be done in conjunction with a Monte Carlo simulation of the primary electron beam trajectories and energy dissipation inside the growing deposit and the substrate. Here, (*x,y,z*) are points in 3D space, that are on the deposit (substrate) surface at any given time *t*. Relevant model parameters are the precursor flux density *Φ* and sticking coefficient *k*, the average precursor residence time *τ*, the energy-averaged dissociation cross-section *σ*, the local electron flux density *f* and the precursor diffusion coefficient *D*. The essentials of this approach are schematically indicated in Figure 2 [**7**]. It is important to note that electron flux density refers to SEs, as these low-energy electrons are most efficient in precursor dissociation, although all other electrons are contributing as well to a minor extend. Thus, the electron flux density *f* is to be obtained from the Monte Carlo simulation. In order to account for local heating effects, that will accelerate thermal desorption of precursor and, at the same time, increase precursor diffusion speed, the Monte Carlo results are also used in determining the temperature distribution over the growing deposit. This is then combined with appropriate correction of *τ* and *D*, both of which following a temperature dependent behavior. Note that a careful determination of the precursor-specific simulation parameters by means of calibration experiments is mandatory for a reliable simulation of 3D-FEBID growth.

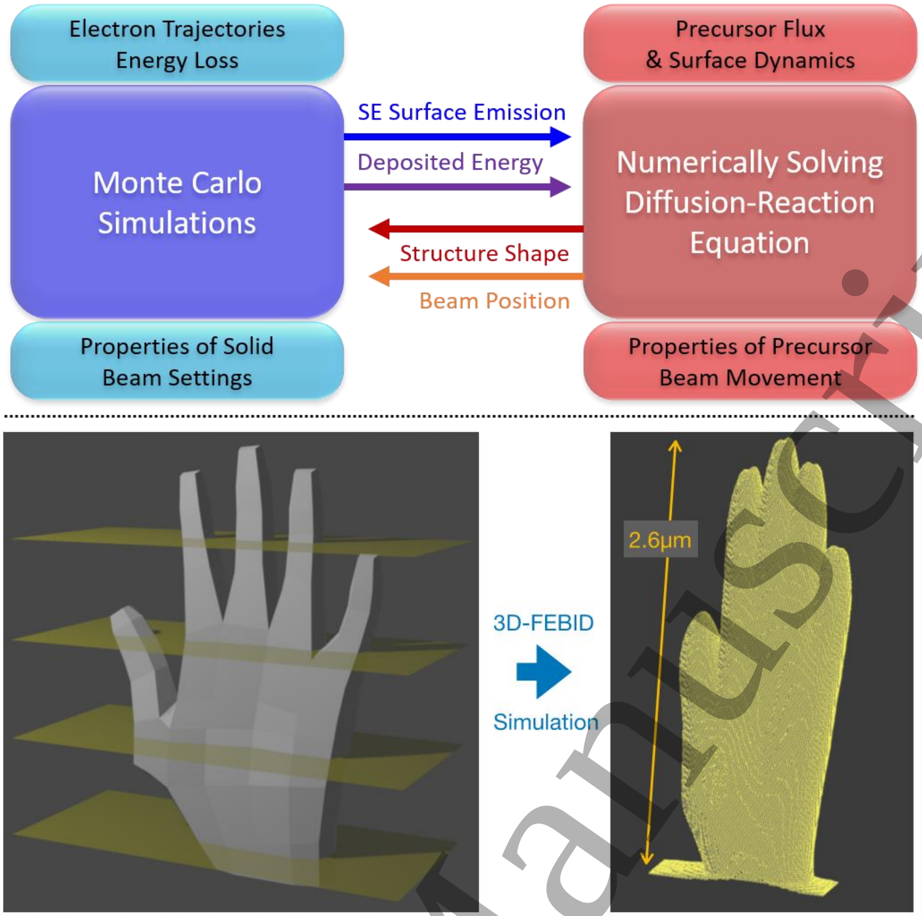

Figure 2.2. (top) Schematic of simulation process flow for 3D-FEBID using reaction-diffusion equation and Monte Carlo simulation. (bottom) 3D test model with exemplary slicing planes as indicated (left). Result of 3D-FEBID simulation using the precursor $Me_3CpMePt$ (right).

2.4 3D Magnetics

In principle, 3D FEBID architectures can be used as scaffolds for subsequent coating via PVD or CVD, which, however, can suffer from spatial inhomogeneity and area selectivity to the 3D structure of interest. That, however, requires further processing, which undermines the idea of direct-write 3D nanofabrication. Consequently, high performance magnetic precursors such as Fe, Co or Co_3Fe can be used for 3DNP, providing metal contents of 80 at.% for the former and >95 at.% for the latter two [**3**]. Together with simulations for preliminary design and reliable transfer, this type of 3DNP paves the way for the fabrication of magnetic nanoarchitectures ranging from free-standing segments to nonlinear helices or nanobridges to highly complex nanoarchitectures. An important area of application concerns scanning probe microscopy based magnetic force microscopy (**MFM**), where even simple, straight pillars of Fe or Co have been used to detect magnetic surface properties with high sensitivity and improved lateral resolution due to sub-10 nm sharp, all-metal apexes [**8**]. The disadvantage of such designs are the narrow pillar diameters down to 50 nm, which can be mechanically challenging during MFM scanning. To compensate for this, a 3D design using Co_3Fe precursors was demonstrated, that shows very high and stable MFM performance (see **Fig. 3A**), even after 8 hours of continuous operation [**9**]. With a view to fundamental research, more complex architectures have been demonstrated, that enable domain wall transport by applying external vector magnetic fields, or curved structures, where geometry-controlled spin structure changes imply an automotion of domain walls. Chirality and anisotropy can not only improve the dynamics of the domain walls but also induce chiral spin states or spin textures with new spin topologies. Demonstrated concepts focused on, e.g., different torsion/curvature of individual helices [**10**], spatially separated double helix strands with different intra-structural properties for magnetic field tuning [**11**] or tightly packed double helices with changing handedness to change chiral spin states, as shown in **Fig. 3B** [**12**]. By applying even more complex 3D designs, artificial lattice structures can be created that are neither provided by nature nor are accessible through chemical synthesis. **Fig.3C** shows a small array of free-standing Co_3Fe nano-cubes that were exposed to external magnetic fields to study the switching behavior of individual branches and the effects on the stray magnetic field for the fundamental understanding of such artificial systems in which frustrated magnetic interactions occur at the branch intersection points [**4**]. In another example, 3D magnetic designs were used to realize a magneto-mechanical nanoactuator concept (see **Fig. 3D**) that replaces electrically driven actuation by external magnetic fields [**13**]. The common element of the mentioned applications is the higher dimensional character in combination with nanoscale elements, enabled by this type of 3DNP.

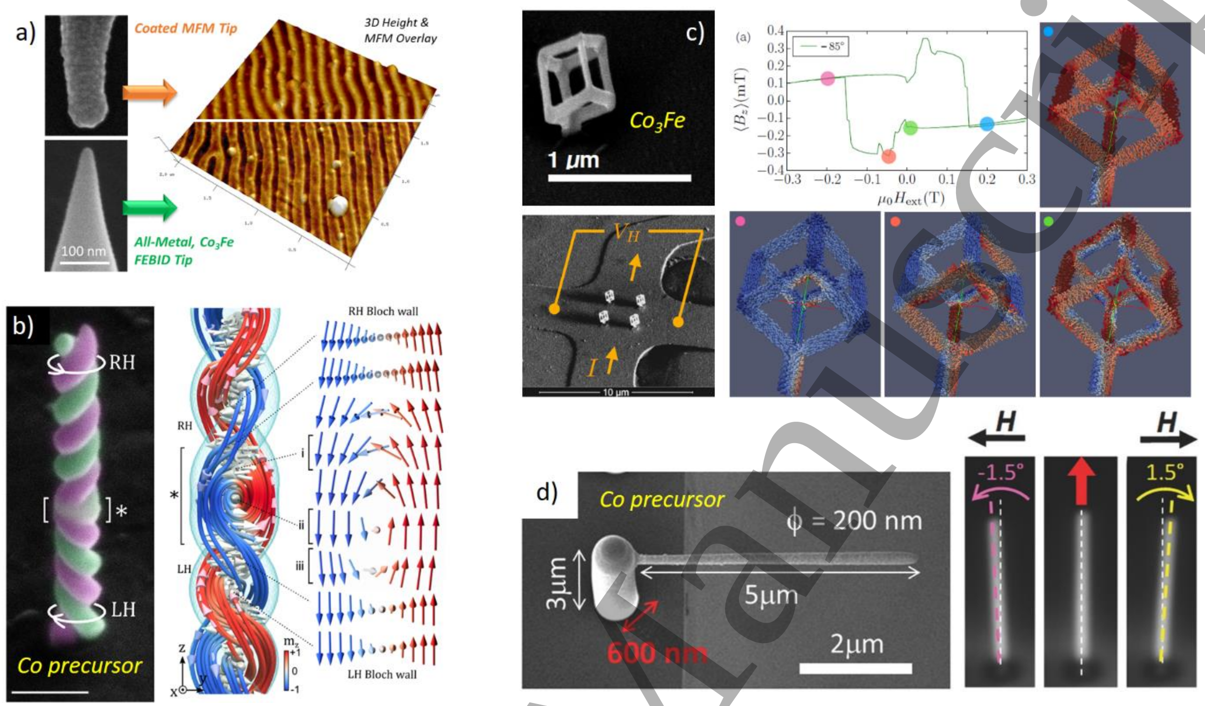

Figure 2.3. (a) SEM comparison of a commercial (top) and a 3D-FEBID (bottom) MFM nanoprobe. The associated MFM signals are shown on the right, illustrating the strongly improved performance due to the sharp all-metal character.[9] (b) Colored SEM image of a close-packed Co double helix (left) with opposite chirality, complemented by micromagnetic simulations (center) revealing the opposite chirality with antiparallel magnetic alignment, as shown in more detail on the right by a RH to LH Bloch wall transition (adapted with permission from Sanz-Hernández et al., ACS Nano 14, 8084 (2020) [12]. Copyright 2020 American Chemical Society). (c) shows a complex Co_3Fe nano-cube (top) further placed on a micro-Hall sensor (bottom) that enables magnetic measurements, complemented by simulations to study the switching behavior of individual branches in response to the external field (right) [4]. (d) shows a composite nano-actuated magneto-mechanical system in a SEM top view (left), which bends left and right in response to an external magnetic field (right), therefore representing the basic element of a magneto-mechanical nanoactuator [13].

2.5 Summary and Outlook

During the past decade, 3D-FEBID has evolved into a reliable technology with unique capabilities due to its direct-write nature and 3D capabilities with nanoscale resolution. Although the technology itself is already well advanced, a remaining challenge is the availability of optimized precursor materials to enable greater flexibility and faster processing.[3] However, it is worth noting that the available Co and in particular the Co_3Fe precursor, remains highly effective for magnetic applications even after fabrication, although efficiency still poses a minor bottleneck for large area processing. The latter also becomes highly relevant when aiming on industrial applications where timely throughput is of great importance. In this context, the introduction of multi-beam instrumentation is highly desired, which can be seen as another remaining challenge in this field. Among the various precursor materials, magnetic 3D nano-objects are of great interest because this type of 3DNP not only enables fundamental research such as spin textures, magnetic dynamics or topological properties, but also offers possibilities in terms of real-world applications including data storage, manipulation or information transport. The latter aspects are elaborated for 3D structures in a recent review by Reisecker et al. [14] together with a general view on the current status of 3D FEBID related applications and remaining challenges.

Acknowledgements
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3 – 3D suspended YIG structures

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3.1 Status

Hybrid magnonics and its application in quantum science kicked-off as an interdisciplinary research field about one decade ago [1]. The fabrication of 3D self-sustained low-loss yttrium iron garnet (YIG) structures, pioneered by researchers from the Martin Luther University Halle-Wittenberg [2], provides a new scalable platform for hybrid magnonic cavity systems. Here, we briefly discuss optical photon-magnon coupling in suspended YIG microdisks and magnon-phonon coupling in micromechanical bridges or cantilever resonators. Compared to 3D YIG spheres with a diameter of 0.3 – 1 mm studied thus far [3-6], on-chip suspended YIG microdisk cavities fabricated at Aalto University (Fig. 3.1(a)) provide much better spatial overlap between TE and TM polarized optical whispering gallery modes (WGMs) and spherically symmetric magnon modes (Fig. 3.1(b)) because of reduced size and dimensionality, which could enhance the optomagnonic coupling strength ($g_{\text{o-m}}$) by several orders of magnitude. Favourable scaling of $g_{\text{o-m}}$ is supported by the rich WGM spectrum of micron-sized YIG cavities, enabling frequency matching and angular momentum conservation between various TE/TM mode pairs and magnon excitations at the triple-resonance condition (Fig. 3.1(c),(d)). After establishing efficient optomagnonic coupling, the YIG microdisk cavities could be used for microwave-to-optics quantum transduction, as strong coupling between magnons and microwave photons has been demonstrated already [1]. This would open a route to the integration of superconducting and optical quantum technologies.

On the other hand, YIG is a very rigid material that promises high quality factors for mechanical oscillators. With bridge-type free-standing mechanical resonators in the micron regime, the resonance frequencies even of the fundamental vibrational modes can easily reach hundreds of MHz or even the GHz [2] regime where strong coupling to magnon modes by magnetoelastic coupling is possible.

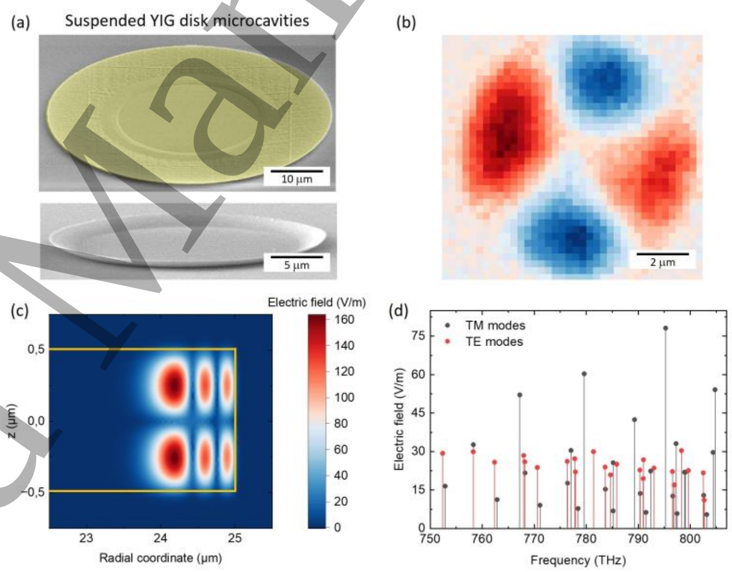

Figure 3.1. (a) Two suspended YIG disk microcavities, one with a 'disk on a stem' structure (diameter of 50 µm) and one with a suspended edge structure (diameter of 25 µm). The YIG disks are 150 nm thick, and their edge is lifted ~1 µm above the GGG substrate. (b) Super Nyquist-sampling magneto-optical Kerr effect (SNS-MOKE) image of a radially symmetric magnon mode in a suspended YIG disk microcavity with a diameter of 7.5 µm. (c) COMSOL simulation of the electric field distribution of a WGM mode in a YIG disk microcavity. (d) WGM spectrum for TM and TE polarized modes in the same cavity.

3.2 Current and Future Challenges

One of the challenges in using 3D self-sustained YIG microdisks as optical cavities for WGMs is the roughness of the disk edge, as it affects the WGM Q factor. In photonics, large Q factors (~10^6) have been reported for suspended silica [7] and AlGaAs [8] disk microcavities with $\Delta r < \lambda/50$. This translates to a maximum allowed edge roughness of ~8 nm and ~30 nm in experiments with blue light and telecom c-band radiation. Another consideration relates to the spatial overlap between the optical and magnon modes. While the spatial overlap between the uniform Kittel mode and WGMs is already much improved in YIG microdisks compared to mm-sized YIG spheres, the use of higher order magnon modes or magnetostatic modes with nontrivial spin textures could maximize the overlap with WGMs at the circumference of the YIG disk even further.

The realization of magnon-phonon coupling requires the careful optimization of the oscillating part as well as of the connection to the substrate. While large YIG spheres are typically mounted on a fiber [9] the suspended YIG structures are directly grown on the substrate and the connecting parts are the most likely leakage paths for vibrational energy. Fortunately, the fabrication technology developed in Halle [2] allows for a large degree of flexibility in device design (Fig. 3.2(a)). In terms of surface roughness, the requirements for mechanical oscillators are even more stringent than for optical devices. Ideally, the ratio between the surface and volume should be minimized and as the surface and edge roughness may be detrimental for the resonance linewidth.

3.3 Advances in Science and Technology to Meet Challenges

Recent advances in the fabrication of 3D self-sustained YIG structures have enabled new research on hybrid magnonic cavity systems. The favourable properties of these 3D systems such as low magnetic damping, on-chip scalability, and flexible tuning of the resonances through cavity design or an external magnetic field are expected to provide higher coupling strengths between magnon, optical, and mechanical excitations. In the advanced fabrication process, the self-sustained 3D YIG structures are patterned by e-beam lithography, room-temperature YIG film growth, lift-off, and crystallization of the YIG structure by annealing in oxygen atmosphere [2]. The challenge of edge and surface roughness has also been addressed recently, leading to promising results. Using a wet etching process after the annealing step, edge seams are removed and smooth edges and surfaces are achieved (Fig. 3.2(b)), that, with a roughness of 10 nm or less at least satisfy the requirements for optical microcavities. Also, in all-YIG mechanical microresonators a proof-of-principle could be demonstrated (Fig. 3.2(c),(d)) by the group of Hans Hübl who measured mechanical oscillations using interferometry [10] on micron sized YIG cantilevers fabricated in Halle.

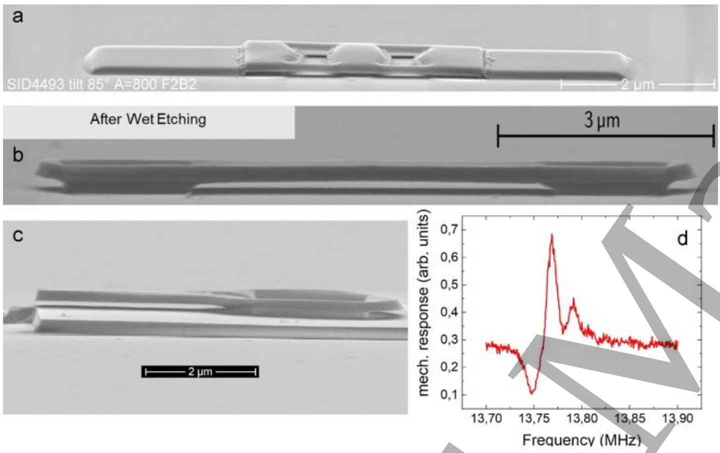

Figure 3.2. (a) A 'trampoline' YIG resonator where a large free-standing mass is coupled by thin connectors to the posts on the substrate. (b) YIG bridge optimized by wet etching for extremely smooth surfaces and edges. (c) YIG cantilever and (d) mechanical resonance of the cantilever measured by interferometry. The quality factor is >500.

3.4 Concluding Remarks

Although a young research field, the development of 3D suspended YIG structures has already achieved a state beyond the mere proof of principle. Structures have been fabricated that theoretically fulfil the requirements for optical photon-magnon coupling and the first mechanical resonances have been measured though at relatively low frequency.

Acknowledgements

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4 - Characterization of 3D nanomagnets

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4.1 Status

3D nanomagnets occur in the form of spatially confined 3D nanostructures and magnetic solitons, i.e., topological spin textures, in solids. In either case, the structural and chemical order governs magnetic and magneto-transport properties [1] whose characterization challenges conventional techniques based on magnetometry and electronic transport. In this section, we outline a selection of emerging advanced characterization techniques that combine high sensitivity with spatial and temporal resolution. These include magneto-transport and microwave spectroscopy for probing individual 3D nanomagnets [Fig. 4.1] and electron holography for visualizing the magnetization vector field [Fig. 4.2].

The capability of magneto-transport for inferring the magnetization configuration in individual 3D nanomagnets was demonstrated on the example of 3D nano-cubes and nano-trees using a high-resolution micro-Hall magnetometer [Fig. 4.1(a)] based on a two-dimensional electron gas in AlGaAs/GaAs heterostructures [2]. A particular field-temperature sweep allowed to tune the magnetic configuration of single elements towards a switching instability. In a different study [3], microwave excitation antennas in proximity to 3D nanovolcanoes enabled the examination of collective spin precessions (spin-waves) in individual nanostructures [Fig. 4.1(b)]. The inhomogeneous internal magnetic field in the nanovolcano leads to a confinement of the magnon eigenmodes within its crate, resulting in a 30% smaller footprint for the same normal mode. Very recently, Brillouin light scattering – ferromagnetic resonance (BLS-FMR) was used to compare the thermal magnon spectra for plank and bumped magnon waveguides [Fig. 4.1(c)] [4]. The geometrically induced non-uniformity of the internal magnetic field gives rise to higher-order spin-wave modes that are absent in planar conduits, solely exhibiting the Kittel-like mode (dashed line in the BLS-FMR spin-wave spectrum in Fig. 4.1(c)).

Further insights into the magnetization configurations and their dependence on curvature, shape, interfaces, inhomogeneity, and defects, also affecting dynamic properties, require direct visualization of the magnetization. In the context of magnetic phase contrast imaging of non-collinear spin textures, off-axis [5,6] and in-line [7] electron holography with transmission electron microscopy (latter also known as Fresnel/Lorentz mode imaging) have reemerged [Fig. 4.2]. Their superior spatial resolution and sensitivity to two components of the magnetic induction complement vector field x-ray tomography (section 5), x-ray magnetic microscopy, scanning electron microscopy with polarization analysis, spin-polarized scanning tunneling microscopy, and nitrogen-vacancy center microscopy.

Aside from advances of individual techniques, a multimodal characterization is highly desired to ensure an efficient and conclusive development of 3D nanomagnet devices. This will also require operandi and interfacing comparative studies of electronic transport, magnetometry, and microwave and optical spectroscopy with magnetic imaging.

4.2 Current and Future Challenges

An interesting application of wire-frame 3D nanomagnets, such as nano-cubes, nano-trees, and nano-networks, is their use as building blocks for artificial lattice structures exhibiting highly complex magnetization distributions (section 7). It might be possible to suppress the occurrence of curling magnetization states in zero field if the junctions between the edges of the building blocks can be made from non-magnetic material. Furthermore, to ensure a ground state with uniform magnetization, the diameter of the roughly cylindrical magnetic edges should not exceed about five to seven times the exchange length, i.e., about 20 to 30 nm for Co-Fe. Reaching such a precision while guaranteeing structural, chemical, and magnetic uniformity has remained a fundamental challenge. While primarily being a nanofabrication challenge, the magnetization switching involving rather complex configurations, such as magnetic vortices or magnetic pinning, poses strict requirements on the sensitivity and spatial resolution of characterization techniques. The expansion of 3D nanostructures and corresponding reduced inductive coupling efficiency of microwave excitation/detection antenna compared with planar thin films limits applicability of microwave probing of complex magnetization configurations. To this end, the sensitivity of FMR can be substantially improved using microresonator loops around the nanomagnet or utilizing its symmetry to tailor the magnon eigenfrequencies. Further limiting factors for studies of curvature and shape effects on the dynamics of spin excitations in 3D nanomagnets are spin-correlation and spin-wave decay lengths of few micrometers.

Isolating intrinsic from extrinsic properties is essential to synthesis/nanofabrication, basic sciences, and eventual device applications and only possible by correlating magnetic properties with structural and electronic characteristics. Providing sufficient sensitivity and spatial resolution beyond those offered by advanced x-ray techniques (section 5) has remained a challenge. Phase contrast imaging with off-axis [5] and in-line [7] electron holography and electron tomography [6] has emerged as one of the leading techniques to visualize the magnetization configuration. Examples include the remanent magnetization in 3D nano-networks [Fig. 4.2(a)] [5] and topological magnetic states in nano-cones [Fig. 4.2(b)] [6]. However, investigations of magnetization switching, dynamics, and phase transitions are currently challenging, if not impossible, due to technical limitations. These studies demand the application of a tunable magnetic field fixed to the sample coordinate system, a temporal resolution ranging from the second down to nanosecond timescale, and variable temperatures, in some cases, down to ~10 K. Overcoming these limitations would simultaneously enable the visualization of spin and charge distributions in other classes of materials, e.g., topological insulators and superconductors.

4.3 Advances in Science and Technology to Meet Challenges

Spatially resolved microwave spin-wave spectroscopy was demonstrated with a coplanar waveguide moving relative to the substrate containing a series of magnetic nanodisks [Fig. 4.1(b)]. Thus, its extension for 3D nanomagnets of cylindrical symmetries, e.g., nano-volcanoes and pyramids, can be anticipated [8]. The requirement of close proximity of the sample under study to an inductive antenna in conventional microwave spectroscopy may be lifted by switching to spin-transfer torque. This approach necessitates (spin) current injection and imposes even stricter requirements on 3D nanoprinting and nanofabrication, which however will benefit structural, electronic, and magnetic properties and eventual device applications.

An interesting approach to study spin textures, such as magnetic vortices in magnetic disks, has recently been demonstrated using ferromagnet/superconductor heterostructures [Fig. 4.1(d)] [9]. The displacement of the magnetic vortex core from the center of the disk affects the supercurrent interference pattern in a Josephson device. Expanding this approach to superconductors in the Shubnikov phase may provide means to derive the magnetization configuration in 3D nanomagnets via stray-field coupling to the Abrikosov vortices. In return, the configuration of the stray field emanating from the 3D nanomagnet should allow for controlling the Abrikosov vortex dynamics and enhancing the current-carrying capability of the superconductor.

Cryogenic, time-resolved investigations down to a few Kelvins will require improved mechanical and thermal stability of aberration-corrected transmission electron microscopes equipped with sensitive single-electron detectors and multifunctional sample holders. The increased data volume and more complicated data acquisition compared with conventional transmission electron microscopy will benefit from an automated optimization, recording, alignment, and reconstruction. The commonly used transport-of-intensity equation [7] to reconstruct the electron phase from the electron intensity is a robust, fast approach but falls short in spatial resolution to the Gerchberg-Saxton exit wave reconstruction [10] as experimentally and numerically demonstrated [Figs. 4.2(d),(e)] [7]. This difference in information becomes essential for imaging the magnetization configuration in materials with small saturation magnetization, surface roughness or inhomogeneities requiring large defocus values or correlation between structural and magnetic properties.

4.4 Concluding Remarks

The profound impact of structure, i.e., shape, curvature, interfaces, and defects, on electronic and magnetic properties of 3D nanomagnets requires the development of sensitive multimodal/operandi techniques to probe static and dynamic properties at variable temperature, electromagnetic fields, etc. Correlating structural nanoscopic features with electronic and magnetic microscopic and macroscopic quantities will be key to advancing understanding and synthesis capabilities. This pertains to the magnetization configurations, phase transitions, and spin excitations. Four specific avenues worth to pursue are (i) enlarging spin correlation and spin-wave decay lengths in 3D nanomagnets (synthesis), (ii) improving (spin) current injection into 3D nanomagnets (synthesis/nanofabrication), (iii) enhancing coupling of 3D nanomagnets to the excitation/detection antennas while providing spatial information (nanofabrication/microwave engineering), and (iv) leveraging phase contrast imaging for operandi characterization (characterization/data analysis).

Acknowledgements

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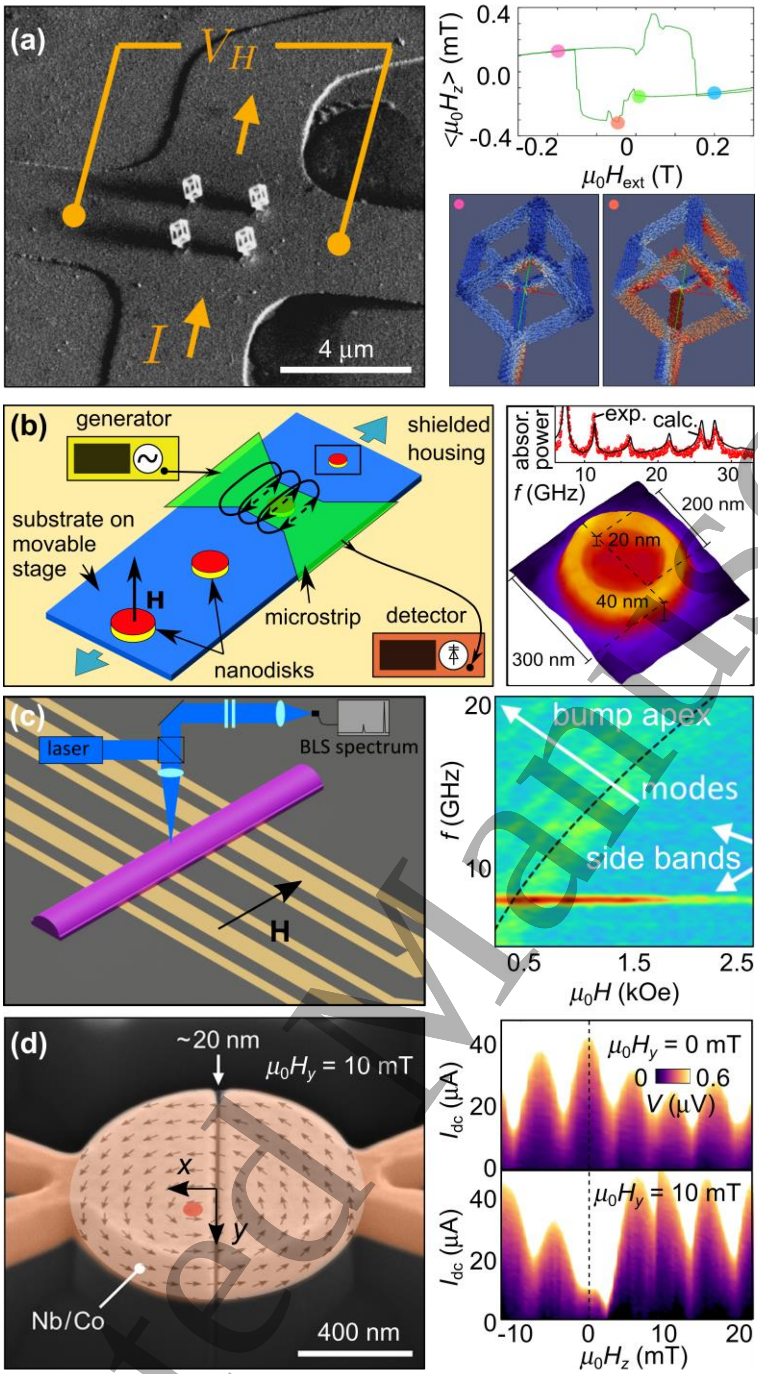

Figure 4.1. (a) SEM micrograph of a Hall cross with a 2x2 array of nano-cubes on its surface in the Hall measurement configuration next to its numerically modeled magnetic hysteresis loop and magnetization configurations for the external magnetic field applied at an angle of 85° relative to the surface normal of the Hall sensor. Adapted from [2], CC BY. (b) Left: Concept of spatially-resolved perpendicular spin-wave spectroscopy for a series of nanomagnets which are individually positioned close to a microwave excitation antenna. Adapted from [8], CC BY. Right: Exemplary spin-wave eigenmode spectrum of a 3D Co-Fe nanovolcano in an out-of-plane magnetic field. The nonuniform internal field of the nanovolcano leads to a stronger confinement of the magnon eigenmodes within its crater. Adapted from [3], CC BY. (c) Schematic of a Co-Fe spin-wave conduit (plank with a bump along its axis) revealing a much richer BLS-FMR magnon spectra in comparison with a reference planar conduit. Adapted from [4], CC BY. (d) SEM image of a Josephson device (Nb/Co disk in which Nb electrodes are separated by a trench, forming a Co weak link) with the superimposed image of calculated spin texture for $\mu_0 H_y$ = 10 mT. The vortex core (red dot) is shifted from the disk center and this affects the supercurrent interference pattern. Adapted from [9], CC BY.

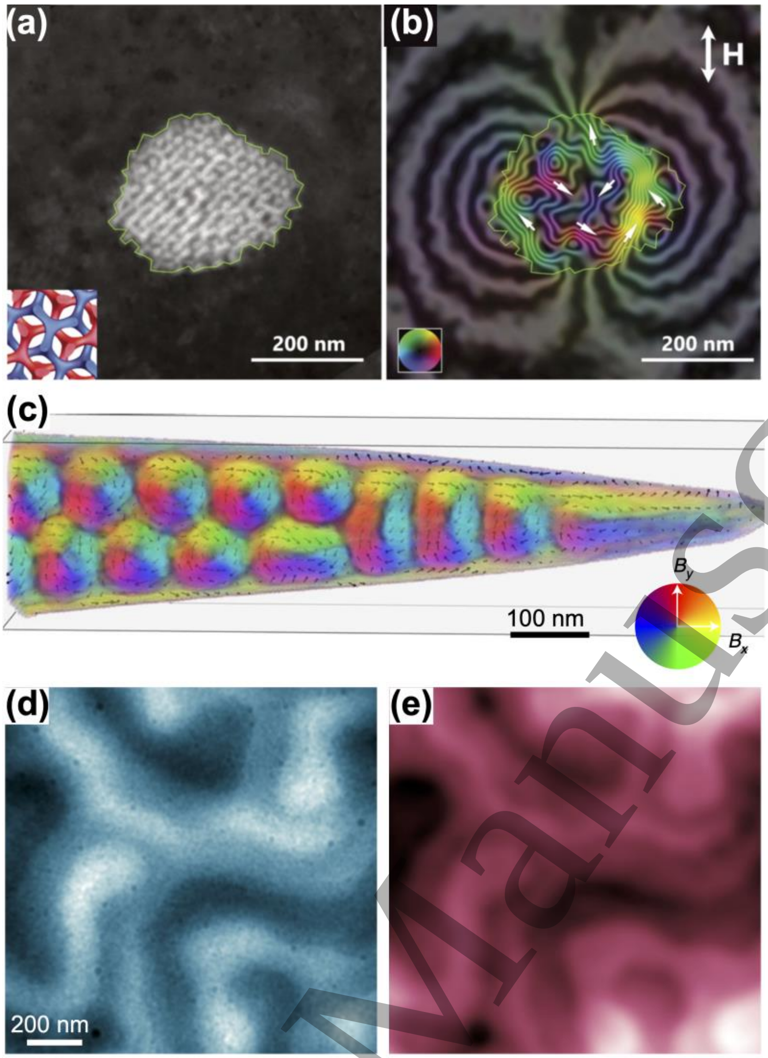

Figure 4.2. Phase contrast imaging with off-axis and in-line electron holography using aberration-corrected transmission electron microscopy. (a) Structure and (b) magnetic induction of 3D gyroid network visualized with off-axis holography. Adapted with permission from [5]. (c) Magnetic induction of topological magnetic states in helimagnetic FeGe nano-cones obtained with holographic vector field electron tomography. Adapted with permission from [6]. (d,e) Electron phase of ferrimagnetic domain walls in amorphous films with perpendicular magnetic anisotropy reconstructed with (d) iterative (Gerchberg-Saxton) and (e) non-iterative (transport-of-intensity) phase retrieval algorithms. Adapted with permission from [7].

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5- 3D nanomagnetic imaging

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5.1 Status

The development of three-dimensional magnetic systems brings opportunities for both new physics and applications. The third dimension can be introduced in two ways: by the extension of the magnetisation vector field to three-dimensional space, and by the introduction of three-dimensional confined geometries. In the first case, the greater degrees of freedom that exist with three-dimensional spin configurations give rise to not only for more complex configurations – with the existence of Bloch point singularities allowing for transformations of topology in space – but also for truly three- dimensional topological textures to exist, such as knot-like magnetic hopfions.
For the second case, patterning magnetic materials in three dimensional geometries offers a route to tailoring the properties of a system via curvature, as well as torsion-induced anisotropy and chirality. In this way, not only can the energy landscape of magnetic textures be tailored, but the breaking of symmetry can lead to exciting dynamics including non-reciprocity and ultra-fast domain wall propagation.

While, initially, the move to three dimensions was mainly driven by theoretical simulations and models, advances in both fabrication and characterisation in recent years have facilitated their experimental realisation. In particular, with the recent development of three-dimensional nanoimaging capabilities, it is now possible to directly map complex three-dimensional configurations and their response to external stimuli, opening a route not only to confirm theoretical predictions – such as the surrounding configuration of Bloch point singularities [1–3] – but also to make unexpected observations of features such as magnetic vortex rings and torons.

When we refer to three-dimensional nanoimaging, the principles of which are presented in Fig. 5.1, we consider tomographic techniques where the three-dimensional vector field – be it the magnetic induction $\vec{B}$ (electron microscopy) or the magnetisation $\vec{m}$ (X-ray microscopy) – is reconstructed in three-dimensional space. The basic principle of tomographic techniques involves the measurement of high spatial resolution two-dimensional transmission projections of the vector field for many different orientations of the sample (typically multiple tomographic rotation axes) with respect to the probing beam. This is followed by the use of an appropriate reconstruction algorithm [4] to recover the three-dimensional magnetic configuration. So far, 3D magnetic nanoimaging has been achieved with both electron microscopy and X-ray microscopy, as summarized in Fig. 5.2.

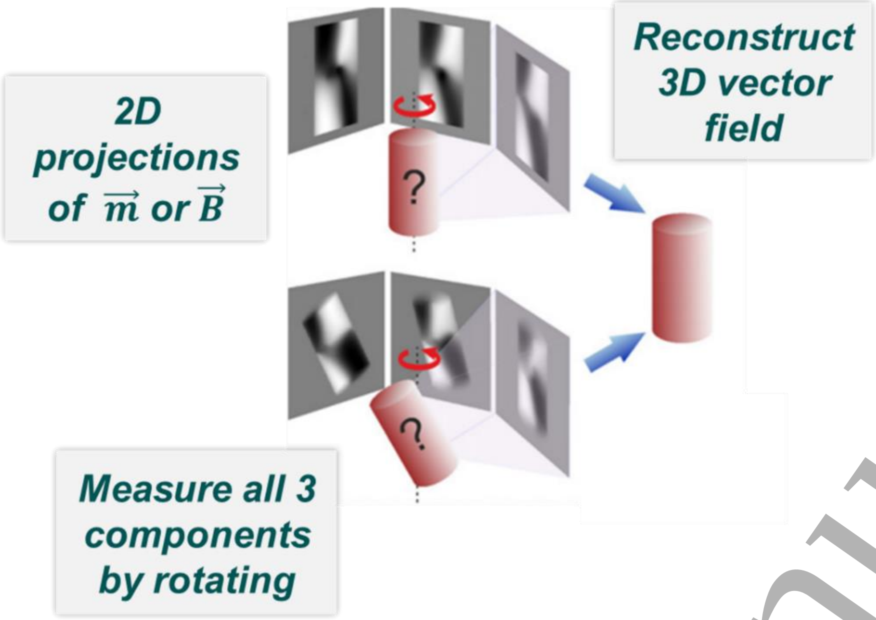

Figure 5.1. The principles of 3D magnetic nanoimaging. Two-dimensional transmission projections of the magnetic induction $\vec{B}$ (Electron imaging) or the magnetisation $\vec{m}$ (X-ray magnetic circular dichroism imaging) are acquired for many different orientations of the sample with respect to the probe beam. Sufficient orientations should be acquired to ensure that all components of the vector field are sufficiently sampled. The final three-dimensional magnetic induction or magnetisation vector field is then reconstructed using a dedicated reconstruction algorithm. Schematic reproduced with permission from [4].

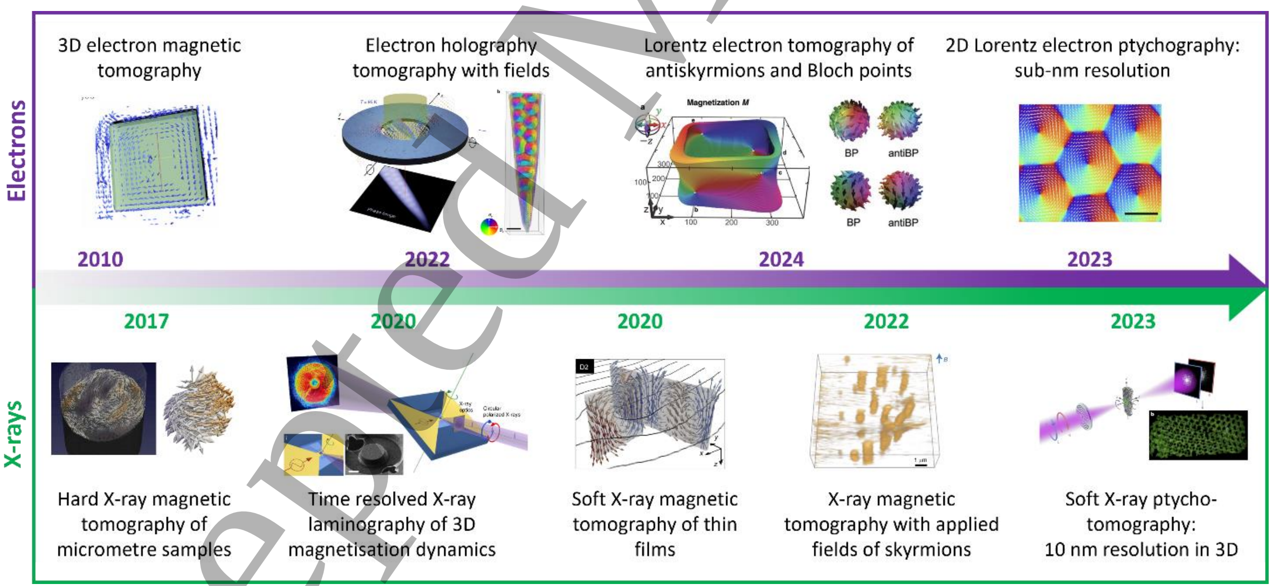

Figure 5.2. Developments in 3D magnetic nanoimaging, with milestones in both electron (upper panel) and X-ray (lower panel) 3D imaging shown over the years. Images reproduced with permission from [1,2,3,5,6,7,9,10,12].

3D magnetic nanoimaging with electrons

With transmission electron microscopy, high spatial resolution projections of the in-plane components of the magnetic induction $\vec{B}$ are obtained, with spatial resolutions down to single-digit nanometres. Due to the limited penetration depth of electrons, sample thicknesses are constrained to approximately 200 nm. By measuring the projections about two independent tomographic rotation axes, it is possible to recover the three-dimensional magnetic induction. 3D magnetic nanoimaging with electrons has been achieved with both Lorentz microscopy [5,6] and electron holography [7], reaching spatial resolutions of less than 10 nm. With these electron-based imaging techniques, the 3D nanoscale configurations of both skyrmions [7] and antiskyrmions [6] have been elucidated.

3D magnetic nanoimaging with X-rays

Synchrotron X-ray magnetic tomography exploits X-ray magnetic circular dichroism (XMCD), which directly probes the element specific magnetisation $\vec{m}$ of a material. X-ray magnetic tomography was first achieved with hard X-rays, where the higher penetration depth provides access to the internal magnetic configuration in micrometre-thick magnets.[1] Due to the weak X-ray magnetic dichroism in the hard X-ray regime, investigations were initially limited to rare earth materials. The subsequent implementation of magnetic tomography with soft X-rays extended imaging capabilities to non-rare earth materials [2], albeit limited to thicknesses on the order of hundreds of nanometres. In this way, spin textures such as Bloch point singularities [1–3] have been observed, as well as the configuration of patterned 3D nanomagnets. [3,8]

5.2 Current and future challenges

While in the last years there has been remarkable progress in 3D magnetic nanoimaging with both electron and X-ray microscopy, the next years will be critical in determining the successful transition from proof-of-principle demonstrations to useful and effective workhorses for the wider magnetic community. To this end, we identify the following challenges:

The development of sample environments for *in-situ* studies, improvements in spatial resolution and speed of acquisition, and making the techniques more accessible to the wider community.

Applying stimuli such as heat, strain, electric and magnetic fields, and currents, during a 3D magnetic nanoimaging measurement is challenging, as it requires the sample environment to rotate with the sample under investigation. First demonstrations of 3D imaging with *in-situ* magnetic fields have been performed with X-rays, by introducing an electromagnet into a reduced "scalar magnetic tomography" experiment [9], and electrons, by integrating a thin permanent magnet into the sample holder.[7]

Dynamic "4D" nanoimaging of the MHz and GHz dynamic response of the magnetisation to an AC magnetic field has been achieved with the implementation of an alternative sample geometry: magnetic laminography, in combination with pump-probe measurements. Harnessing the picosecond time structure of synchrotron X-rays, time resolved 3D imaging was achieved, revealing the buried motion of magnetisation textures and spin waves. [10,11]

With the development of new measurement geometries and sample holders, *in-situ* studies become possible. However, beyond the technical challenges, a major limitation that remains is the acquisition time required for a high-quality dataset, which is still on the order of several hours or days, even for samples with high magnetic contrast. For synchrotron X-ray beamtimes, where available measurement time is typically limited to a few days, extensive measurements are currently not possible. As a result, progress must be made to make the experiments faster, and to explore more efficient measurement protocols.

The speed of acquisition also impacts the achievable spatial resolution of the 3D nanomagnetic imaging techniques, which is typically related to the speed at which statistically meaningful data can be acquired. This question of obtaining sufficient data in a suitable time also impacts the variety of samples that can be measured. So far, there exist two main regimes in terms of the sample volume that can be probed: the imaging of thin samples of almost arbitrary composition (up to ≈ 2-300 nm in thickness) with electrons or soft X-rays, or the imaging of rare-earth containing thick samples (≈ 1-10um) with hard X-rays. However, the imaging of thicker non-rare earth samples has so far been limited by the very weak signals at the transition metal K edges in the hard X-ray regime, limiting the applicability of 3D magnetic nanoimaging to specific materials and thicknesses. Recent implementations of X-ray coherent imaging, making use of the pre-edge phase XMCD, extend the imaging of transition metal-based systems by an order of magnitude to micrometre-thicknesses [13], opening the study of, for example, thick helimagnets and giant magnetofossils. [13]

5.3 Advances in Science and Technology to Meet Challenges

Since the first demonstrations of 3D magnetic nanoimaging, there have been continuous advances in experimental capabilities, with the extension to a variety of electron and X-ray techniques, and the development of sample environments.

To make magnetic 3D imaging a versatile tool for the wider scientific community, *in-situ* capabilities should be expanded. One priority should be to drive an ambitious project for a dedicated instrument with cryogenic capability as well as the possibility to apply in situ magnetic fields up to 1 T. The ability to freely sweep temperatures and fields will allow for phase diagrams of materials with competing interactions or antisymmetric exchange to be explored, in which 3D topological textures are predicted to be stable. Indeed, the ability to precisely sweep both the temperature and the magnetic field has been key for the creation of both skyrmion and hopfion textures.

Until now, such *in-situ* 3D measurements have involved full tomographic datasets being acquired for each time or magnetic field step, with a certain redundancy in the data. By measuring sparse datasets, where the amount of measured data is optimised, data acquisition times can be reduced by an order of magnitude.

The reduction of long acquisition times is particularly important for synchrotron X-ray imaging. Here, advances in detector technology are crucial, especially for the soft X-ray regime, in which charge couple device detectors (CCD) with slow readout are widely used. The recent development of Low Gain Avalanche Diode (LGAD) sensors would give the advantage of single photon counting pixelated detectors, with the associated reduction in electronic noise, increase in dynamic range and much faster kHz readout speeds.

One of the main opportunities for the future of 3D magnetic nanoimaging, both with electron and X-rays, comes with coherent imaging techniques.

The performance of coherent X-ray methods will significantly improve with the considerable increases in coherent flux associated with next generation synchrotron sources. This will directly translate to higher spatial resolutions and faster measurement times. The increase in coherent flux will especially benefit coherent imaging techniques such as holography and ptychography, for which the spatial resolution is not dependent on optics, but is rather scattering-limited. We envisage 3D X-ray nanoimaging will soon approach single-digit-nanometre spatial resolutions, comparable to that of current electron imaging techniques.

Likewise, coherent imaging techniques offer dramatic improvements in the spatial resolution of electron nanoimaging. Indeed, sub-nanometre spatial resolution imaging of magnetic skyrmions in 2D [12] has recently been demonstrated with Lorentz electron ptychography. If combined with 3D imaging, Lorentz ptychography offers the prospect to be able to image 3D magnetic configurations approaching **atomic spatial resolution**, opening up the possibility for atomic imaging of spin textures such as Bloch point singularities, as well as the ability to resolve antiferromagnetic sublattices.

Finally, a key aspect of 3D nanoimaging is the importance of reconstruction algorithms. Advances in algorithms as well as the use of machine learning offer routes to more efficient, and physics-informed reconstructions. In this way, not only could spatial resolutions be further improved, but 3D nanoimaging could offer nanoscale magnetometry, with the spatially resolved determination of material parameters. Likewise, the computational nature of 3D imaging demands advanced computing facilities: as spatial resolutions improve, and the sizes of datasets increase, appropriate computing resources for the processing and storage of data will be crucial to the continuous development of 3D nanoimaging.

5.4 Concluding Remarks

In the last decade, the inception of 3D magnetic nanoimaging has been an important advance for the experimental investigation of three dimensional nanomagnetic systems. In the coming years, as thefield of 3D nanomagnetism develops, we predict several key opportunities for 3D magnetic nanoimaging:

Magnonics: First demonstrations of time-resolved 3D imaging have offered insight into the 3D dynamics of both magnetic textures and spin waves. Advances in detectors and coherent imaging capabilities could extend 3D time-resolved magnetic imaging to both coherent, driven dynamics and incoherent, thermal dynamics with nanoscale spatial resolution.

Topological defects: With the future flexibility to image a variety of magnetic materials and sample geometries, and the combination with high spatial resolution imaging, the 3D nature of elusive textures such as hopfions and torons, and their dynamics, will become accessible.

Curvilinear systems: High spatial resolution will provide a route to the direct measurement of 3D nanopatterned materials and curved thin films, so providing a way to experimentally determine the influence of curvature on the local magnetic properties.

Spin and orbital moments: As acquisition times improve, the extension to spectral magnetic tomography could allow for the exploitation of the XMCD sum rules, providing a way to resolve both the spin and orbital moments in 3D magnetic textures.

Antiferromagnets: With the increasing interest in both antiferromagnetic textures and curvilinear systems, there is a growing need to extend 3D magnetic nanoimaging to antiferromagnetic materials, which could be achieved by harnessing X-ray magnetic linear dichroism.

Based on the progress in capabilities in the last years, we anticipate significant developments in 3D magnetic nanoimaging in the future, so that it will become an integral tool for the investigation of magnetism and magnetic materials and beyond.

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6- Coupled multilayer systems with ferromagnetic and antiferromagnetic exchange interactions

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6.1. Status

Artificially layered magnetic structures allow control over continuously altering magnetic properties, like anisotropy and exchange, which are usually set and fixed by the specific elemental composition and crystal structure of a material [1,2]. Indeed, in artificially layered systems, it becomes possible to access and explore new energy scenarios and consequently, to vary and to design magnetic properties that usually are difficult to alter in homogenous systems, thus opening up so far unexplored application opportunities. In this regard, two important examples are antiferromagnetically (AF) coupled layered systems with perpendicular magnetic anisotropy (PMA) [1,3], made from metallic magnetic and nonmagnetic materials, and ferromagnetically (FM) coupled graded materials [4], made of a single magnetic layer, in which the material properties are gradually changing along its thickness. Beyond the fundamental interest in such complex artificially layered materials, an understanding of their properties at the nanoscale and their relationship to function, will lead to new applications in a broad range of areas, such as magnetic memory, sensors and actuators, refrigeration, energy storage and neuromorphic computing schemes.
While we focus here in this section on well-established sputter deposited magnetic multilayer systems, another new type of layered magnetic structures, so called 2D-Van der Waals systems, has rapidly evolved in the past decade and also made significant progress to demonstrate a variety of magnetic functionalities [5-8]. Artificially stacked layer sequences of such atomically flat 2D-materials thus will most likely become another pathway to fabricate atomic scale 3D magnetic architectures in the future that could reveal novel or exotic magnetic ordering phenomena. However, in contrast to the sputter deposited magnetic thin film systems presented here, it currently still remains very challenging to alter the magnetic properties of such stacked 2D-Van der Waals materials in a continuous fashion as demonstrated in the two examples presented here.

6.2. Current and Future Challenges

There is an increasing interest in understanding and exploiting artificially layered magnetic structures, since they represent an established and very successful approach to materials design. However, despite recent progress, there are still many unexplored opportunities and remaining challenges. One of the most pressing ones lies in optimizing such systems to control and design 3D magnetic structures and architectures: they could either form naturally by the specific design of the layered structure and the respective magnetic energy balance, or could post-deposition be defined within the thin film plane of the material system by, for instance, focused laser or ion beam irradiation as well as more conventional lithography techniques. Accordingly, the next section discusses two of the most relevant coupled multilayer systems related to the latest advances without necessarily relying on additional post-deposition patterning techniques.

6.3. Advances in Science and Technology to Meet Challenges

6.3.1 Tuning the magnetic energy terms in synthetic antiferromagnets with PMA

Double multi-layered PMA synthetic antiferromagnets (SAFs) (Fig. 6.1a) [1,3], where the competition between AF interlayer exchange coupling (IEC) energy ($E_{AF\text{-}IEC}$), PMA energy ($E_{Ku,PMA}$) and demagnetization energy (E_{demag}) can be continuously tuned, allow the stabilization of novel 3D magnetic structures. While intrinsic antiferromagnets (AFMs) dominated by AF exchange and respective AF-exchange dominated PMA-SAFs, both reveal a reversal via a 1D surface spin flop or an AF inversion followed by a bulk spin-flop [9], the AF-IEC in PMA SAFs can also be easily lowered to the regimes displayed in Fig. 6.1b and 1c [3,10]. In these cases, both FM and AF phases coexist during the magnetic field reversal, in remanent (Fig. 6.1c, inset) or metastable states (Fig.1b, inset bottom right). If $E_{AF\text{-}IEC}$ is reduced below both, $E_{Ku,PMA}$ as well as E_{demag}, (Fig. 6.1c), the remanent state consists of periodic FM stripe and bubble domains with AF ordering still existing within the in-plane Bloch type domain walls [10]. Such recently highlighted structures may be used to encode multi-bit information within one and the same bubble domain that can still be moved along a nanoscale racetrack [10]. Additionally, the AF domain walls inherent in respective aligned parallel stripe domains, thus forming a self-organized magnonic crystal, provide a pathway for guiding spin waves with nonreciprocal propagation characteristics [11]. While these naturally formed mixed FM/AF states themselves can already be very attractive for applications [6], it is also possible to use focused laser or ion beam techniques to locally alter the magnetic energy balance. Such modifications of the local magnetic energy landscape have been demonstrated by deterministically "writing" mixed AF anti-phase domains with 1D FM stripe domains at their boundary (bottom right inset of Fig. 6.1b) [12]. These artificial magnetic 3D states are suggested to create complex 3D magnetic infrastructures for controlling (re-configurable) 2D stray field profiles at the sample surface for data processing and storage via spin wave manipulation [12].

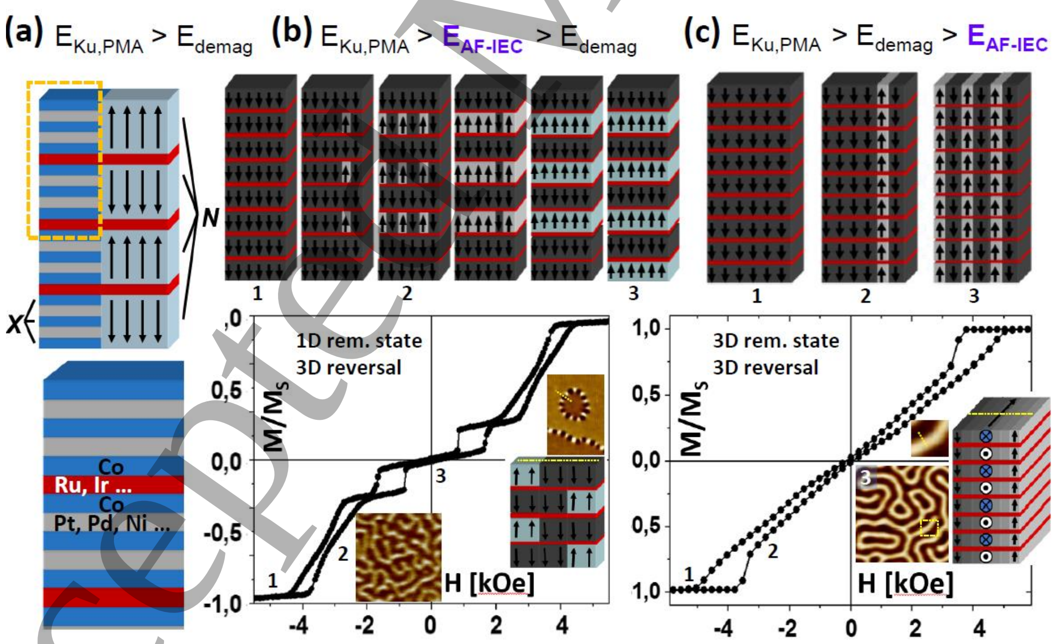

Figure 6.1. (a) PMA-SAFs ($E_{Ku,PMA} > E_{demag}$) based on double multilayer (X, N) magnetic meta-materials, architecture (top), respective material options (bottom). (b,c) Typical magnetic field reversal for (b) $E_{Ku,PMA} > E_{AF\text{-}IEC} > E_{demag}$ [3], and (c) $E_{Ku,PMA} > E_{demag} > E_{AF\text{-}IEC}$ [10]. The top displays schematic illustrations of the reversal from negative saturation to remanence. The bottom shows respective VSM hysteresis loops. Specific reversal stages are labeled for better cross-comparison. Top-down MFM domain images (2μm x 2μm each) are added as insets for reversal state 2 in (b) and for the remanent state 3 in (c) together with a schematic cross-sectional domain wall image [10]. While for case (b) the remanent state is still 1D AFC, for (c) it becomes a complex 3D mixed FM/AF domain structure [3, 10]. The two bottom right insets in (b) display an MFM image together with a cross-sectional scheme of a metastable AF antiphase domain state separated by a 1D FM stripe domain phase as obtained after out-of-plane AC demagnetization [3].

6.3.2 FM phase engineering in exchange-graded thin films

In contrast to the above-described discrete multilayers, the material composition of graded materials changes in a very continuous fashion. Nevertheless, they allow as well to control the associated magnetic properties very precisely [4]. This route to novel material design has recently been taken further with the accomplishment of an effective way to engineer phase transitions and FM phase boundaries [13]. Indeed, formally, a homogeneous magnetic material that exhibits ferromagnetic exchange coupling should exhibit only one single Curie temperature T_C. However, systematic variation of the exchange coupling strength causes graded magnetic materials to behave as if they were composed of virtually independent sub-layers, each exhibiting a "local" Curie temperature T_C^{local} [13,14]. This has a clear impact on the dimensionality of the magnetic states, with the corresponding spin configurations extending not only laterally in the plane, but also vertically along the thickness. Single crystal CoRu alloy films, featuring a V-shaped pre-defined depth dependent exchange coupling (J) profile, as shown in Fig. 6.2(a), are an excellent example of that [15]. Such graded material exhibits a collective versus interface propagating progression of FM phase transitions [15]. Their balance depends on the specifics of the grading profile. In this case, as the samples have a V-shaped depth profile with the largest J in its center, the FM state at T_C forms first in the center as a sort of 2D-FM-coupled interface. By further lowering the temperature, the FM state becomes stable over an ever-larger region of the sample, as schematically depicted in Figs. 6.2(b) - (d), continuously extending into the 3rd dimension perpendicular to the film plane [15]. This reflects itself in the critical exponent, as shown by the temperature dependent magnetization in Fig. 6.2(e), which, in contrast to homogeneous systems, can be tuned in a very wide parameter range [15]. Thus, graded systems with specifically designed exchange coupling profiles allow to give nanometer-scale magnetization state control to materials, with thermodynamics being re-defined on a very short length scale (1-2 nm) [16], in structures that are FM coupled throughout [17]. Consequently, properly designed exchange coupling profiles have the ability, for instance, to design specific temperature dependent characteristics of the coercive field H_C, as depicted in Fig. 6.2(b), which could have a strong influence on the thermal stability and reliability of next-generation devices [17].

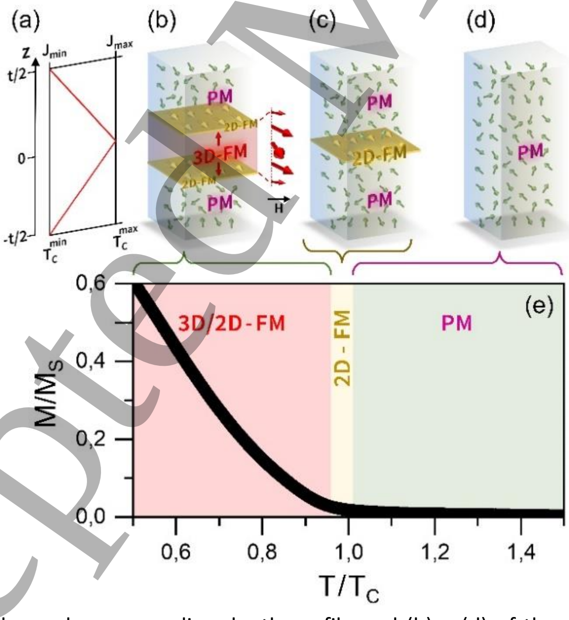

Figure 6.2. (a) schematics of the exchange coupling depth profile and (b) – (d) of the magnetic state profiles upon changing the temperature of a single crystal CoRu alloy film. The arrows refer to local magnetic moments [11]. (e) Corresponding temperature dependence of the normalized magnetization [11].

6.4 Concluding Remarks

The study of complex materials, such as coupled multilayer systems with FM and AF exchange interactions, provides exciting opportunities for gaining an improved understanding of magnetism and magnetic phase transitions. The interplay between multiple degrees of freedom and competing interactions can drive the system into complex mixed phases that further can be manipulated by confinement of the system. Understanding, stabilizing and controlling such new phases should make a broad range of new applications possible.

Acknowledgements

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7- 3D Artificial-Spin Ice Systems

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7.1 Status

Exploring the consequences of frustrated magnetic interactions and associated emergent phenomena via magnetic imaging, until very recently, focused at designing artificial frustrated spin systems strictly confined within the constraints of the two-dimensional (2D) space. Similarly, investigations of the spin dynamics and spin-wave properties in artificial spin-ice networks primarily focused on 2D systems [1]. While this remains a popular endeavour in the field, recent advances in nanofabrication techniques allowed for the exploration of the third dimension as an additional degree of freedom in designing new artificial spin-ice systems and accessing novel properties. This opened up a pathway to introduce three-dimensional artificial spin ice systems [2], [3] that replicate aspects of pyrochlore spin ice, but also go beyond spin ice physics [4]. In addition to their fabrication, magnetic characterization techniques, most prominently soft x-ray ptychography [5], need to be further developed, in order to directly visualize magnetic configurations in such complex 3D magnetic networks. From a statistical physics perspective, one crucial obstacle that needs to be addressed remains the question on how to thermalize such three-dimensional structures, as they all remain frozen, allowing only for field-driven dynamics to be explored, with quasi-three-dimensional artificial spin ices [Fig. 7.1(a), and Fig. 7.1(c)] remaining the only known exception. Beside the fascination for the fundamental statistical physics of magnetic frustration, artificial spin ice systems have also emerged as interesting model systems forming magnonic metamaterials and magnonic crystals. Extending the periodicity into the third dimension reduces the footprint of the lattice, while maintaining the same magnetic material volume contributing to the signal in spin-wave spectroscopy experiments. Furthermore, the transition from 2D to 3D spin architecture allows for the introduction of a whole new range of phenomena, including novel dynamic coupling mechanisms, emergence of new hybrid modes, and the reprogrammable modification of the magnonic bandstructure dynamics as highlighted in Fig. 7.1(b) and Fig. 7.2. For instance, the coupling of a 2D artificial spin-ice system to an extended thin film underlayer was shown to lead to emergent dynamics at either specific wavelengths or intensity modulations peculiar to the modes of the artificial spin-ice elements imprinted in the film [6], see Figs. 7.2(g,h). Even more impressively, it was recently demonstrated that a 3D artificial spin ice comprised of interconnected nanowires arranged on a diamond-bond lattice exhibits collective spin-wave modes with a spatial quantization throughout the network, Figs. 7.2(e,f). [7]. Aside from the high reconfigurability and vast microstate space, 3D artificial spin ice also provides the opportunity to engineer stronger dipolar coupling; hence, maximizing magnon-magnon coupling strength. This was recently realized in a 3D artificial spin ice based on two artificial spin systems separated by an height difference along with a lateral inter-layer offset that breaks the dipolar coupling symmetry between the

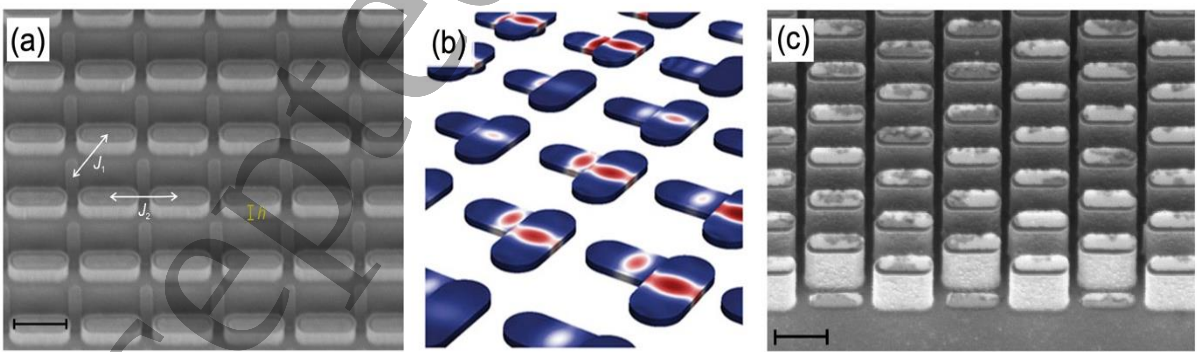

Figure 7.1. (a) Quasi-3D artificial spin ice realized by placing Ising-type nanomagnets on top of pre-etched silicon substrates. The etching depth defines the achieved height offset that allows the tuning of the balance between competing interactions J_1 and J_2 and the realization of macroscopic spin ice degeneracy. It also serves as a basis for novel 3D magnonic crystals (b). (c) Artificial planar triangular antiferromagnet, a concept that could not be realized within the 2D constraints. Scale bar in (a,c) is 400 nm. (a) Adapted from Ref. [3]. (b) Adapted from Ref. [1].

states enabling chiral microstate control and resultant magnonic spectral control [8], see Figs. 7.2(a-d).

7.2 Current and Future Challenges

Most 3D artificial spin ice structures that are being increasingly introduced lack one important ingredient: thermally-driven moment reorientations. Until this obstacle is overcome, the thermodynamics and statistical physics and associated emergent phenomena in these systems will remain an elusive goal in the field. Furthermore, even if a thermodynamic 3D artificial spin ice was introduced, current magnetic imaging techniques simply do not operate at timescales that would allow for real-time characterization of thermodynamic processes in these systems. However, x-ray scattering techniques, for example x-ray photon correlation spectroscopy or Muon spectroscopy could already provide access to such thermodynamics. A major obstacle in the fabrication of "fully 3D artificial spin ice" is that the current approaches are limited to interconnected lattices and to only one unit cell in the third dimension, i.e., the vertical direction [9]. These problems could be circumvented by implementing deposition techniques that are not based on a "line-of-sight" techniques. This means that non-visible surfaces of the structure would also be coated creating "bulk" 3D artificial spin-ice structures. The detection of spin waves in a complex 3D system such as a 3D artificial spin ice bears two key challenges: (1) The 3D nature of the system requires the integration of different levels along the vertical z-directions, making the local detection and excitation of spin waves by electrical techniques difficult. (2) Currently-employed probing methods rely either on radio-frequency (RF) techniques such as spin-wave absorption spectroscopy or magneto-optical techniques such as Brillouin light scattering. RF-based techniques typically detect the collective dynamics of the entire system, while magneto-optical techniques primarily probe the most top layers of the magnetic material. Hence, it is challenging to experimentally access dynamics in the bulk of the structure.

7.3 Advances in Science and Technology to Meet Challenges

Considering that the attractiveness of artificial spin ice systems lies in their function as model systems that allow for the direct real-space visualization of magnetic frustration and as an effective magnonic system with reprogrammable properties, it is of crucial importance that nanofabrication, magnetic imaging and spin-wave sensing technologies to develop hand in hand to deliver the next revolutionary milestone in the field. While scanning probe microscopy techniques such nitrogen vacancy (NV) magnetometry could provide a route in visualizing frozen configurations, after thermal annealing procedures, it is very likely that the next generation of synchrotron light sources will deliver that much needed milestone of directly observing the real-time thermodynamics in a 3D artificial spin ice. On the other hand, NV-center based sensing is expected to enable nanoscale detection of spin waves in 2D and 3D magnonic structures, which could be utilized as a local readout of spin-wave interference information. Furthermore, RF microresonators have successfully been employed to probe spin dynamics in iron-filled carbon nanotubes [10]. These types of detection methods as well as inverse-spin Hall effect-based measurement techniques are compatible with 3D artificial spin ice and could even serve as local readout in magnonic devices.

7.4 Concluding Remarks

In summary, 3D artificial spin ices, while still at an infancy stage, is expected to gradually grow to become a major line of research in 3D nanomagnetism. We anticipate that 3D artificial spin ice will accelerate ongoing efforts in developing energy-efficient functional magnonic applications and enhance neuromorphic computing capabilities based on artificial spin systems. Its emergence will not only lead to the observation of novel phenomena in statistical physics and spin-wave dynamics but will also result in the development and introduction of new fabrication and characterization technologies that will then serve the broader research community.

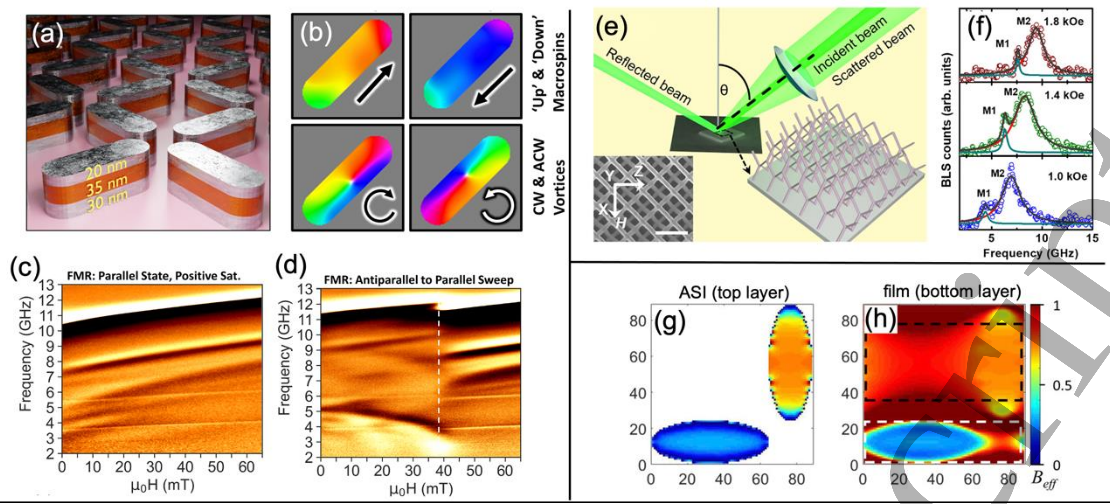

Fig. 7.2: (a-d) Demonstration of ultrastrong magnon-magnon coupling and chiral symmetry breaking in a 3D layered artificial spin ice (ASI): (a) Schematic illustration of array. (b) Four possible macrospin and vortex nanoisland magnetization states. (c) Parallel state exhibiting only the acoustic magnon mode, whereas (d) antiparallel state also shows the optical magnon mode. (c-d) Adapted from Ref. [8]. (e,f) Coherent spin waves in a 3D ASI detected by Brillouin light scattering (BLS): (e) Schematic of measurement configuration with scanning electron microscope image as inset. (f) Various magnon modes detected by BLS. (e,f) Adapted from Ref. [7]. (g,f) Observation of dynamic coupling in a magnetic hybrid system made of an artificial spin-ice structure and an extended NiFe underlayer: ASI mode in the top ASI layer (g) imprints dynamics in the thin film underlayer (h). (g,h) adapted from Ref. [6].

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8- 3D magnetoplasmonics

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8.1 Status

Plasmonics provides a unique approach to confine and enhance electromagnetic radiation at the nanoscale, bringing a huge potential for novel applications, for instance, in energy harvesting, optoelectronics, and photochemistry. To achieve novel functionalities, finding new synergies between plasmonic and other material properties has become increasingly attractive. Magnetoplasmonics, the combination of magnetism and plasmonics, is a promising route to enhance magneto-optical and opto-magnetic functionalities, including ultrafast magnetic phenomena [1]. Until now, the large majority of works in magnetoplasmonics have been focused on flat planar nano-objects and arrangements, namely two-dimensional (2D) systems, such as 2D nanoantennas and crystals. Initial attempts to exploit the third dimension showed that the out-of-plane interaction between metallic nanoantennas stacked vertically (see Figure 8.1a) can lead to (i) the appearance of magneto-optical activity in the purely plasmonic disk and (ii) to both an enhanced and a vanishing (i.e., transparent) magneto-optical response (Figure 8.1b) at specific wavelengths [2]. The additional collective optical and magneto-optical excitations that can be excited in magnetoplasmonic, so far only 2D, crystals, made of such vertically stacked nanoantennas, were later shown to enable (i) superior refractive index sensitivity [3], and (ii) a versatile modulation of the magneto-chiral response of nanoscale systems with enhanced enantio-sensitive functionalities [4]. It has been also shown that 2D magnetoplasmonic crystals made of in-plane geometrically anisotropic nanostructures allow a non-trivial interplay between the directly excited and the magneto-optically induced electric dipoles (see Figure 8.1c), thus providing a broadband control of light polarization and intensity [5]. This concept was later extended, only theoretically, to three-dimensional (3D) ferromagnetic nanoantennas (e.g., pillars and cones), where the combination of resonant and off-resonant localized plasmonic excitations along the three directions in space further allows optical loss control and the boost of the magneto-optical response (see Figure 8.1d) [5].

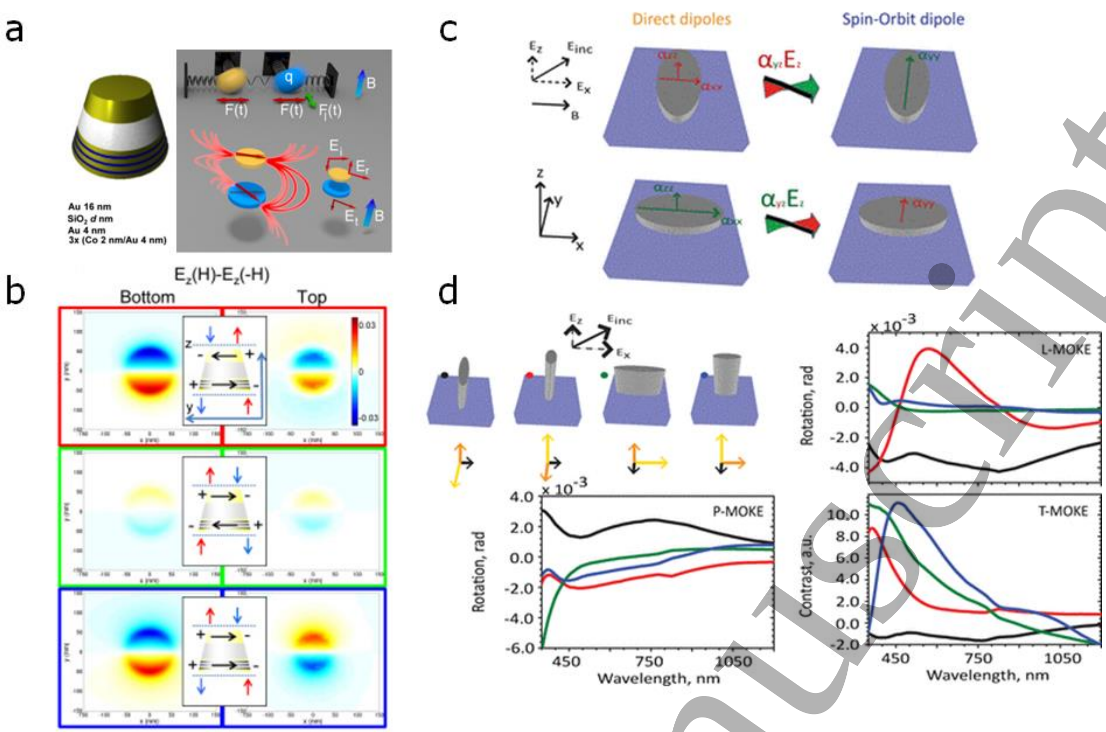

Figure 8.1. (a) Left panel: schematic drawing of the nanoresonators composed of a purely plasmonic Au disk and a magnetoplasmonic Au/Co superlattice disk separated by a dielectric spacer. Top-right panel: spring model representing a two coupled masses system excited by a harmonic force, F(t), along x axis. One of the masses (blue) is charged (q) and a static magnetic field (B) is applied along the z direction, inducing a Lorentz force, $F_l(t)$, along the y direction. The y-movement is transferred to the other mass through the coupling. Bottom-right panel: two interacting electric dipoles, representing two metallic disks, excited by an incident beam polarized along the x axis. One of the disks (blue) has magneto-optical activity and a static magnetic field (B) applied along the z direction induces a rotation of its electric dipole, which is transferred to the other dipole through the interaction. The rotation modifies the polarization direction of the reflected (E_r) and transmitted (E_t) light. (b) Difference of the E_z components for magnetic saturation along opposite directions in the same planes and for the same structure. This difference accounts for the effect of the applied magnetic field: the appearance of a dipole along the y direction for both the magnetoplasmonic (intrinsic dipole) and the plasmonic (induced dipole) disks. In both cases, the components for three different wavelengths labelled with different colours (red, green, and blue frames). At specific frequencies (e.g., see green frame) the magneto-optical response is suppressed due to the destructive interference between the two dipoles. (c) Schematic overview of the interplay of the directly excited (left, resonant α_{xx} and off-resonant α_{zz}) and spin–orbit coupled (right, resonant α_{yy}) nanoantenna modes that all together deliver exceptional light polarization rotation control. Light incidence (p-polarized) and applied magnetic field are marked. (d) Schematic set of 3D nanoantennas (top, left) with highlighted dipolar on (yellow, orange)/off (grey) resonance modes. p-polarized light is always used. Calculated Kerr signals for the nanoantennas in panel in L-MOKE (top, right), P-MOKE (bottom, right), and T-MOKE (bottom, left). Panels (a) and (b) are reproduced from Ref. [2]. Panels (c) and (d) are taken from Ref. [5].

Initial modelling efforts on 3D plasmonic crystals showed the enormous benefits that would arise from their far richer optical band structure and hybridization effects [6]. 3D magnetoplasmonic crystals are expected to show the very same effects, in this case on both the optical and magneto-optical band structures, thereby enabling a far improved and richer magneto-optical response as compared to the present 2D counterparts [7]. In addition, the emergence of unique 3D spin textures occurring in 3D nanomagnetic systems (see Sections 9 and 11) that can extend over distances comparable with the radiation wavelength in the visible and near-infrared spectral range, should enable a superior magnetic control of light.

8.2 Current and Future Challenges

These initial incursions of magnetoplasmonics in the 3D realm mentioned above had already provided a clear hint of the great potential that the exploitation of the third dimension could unleash, and efforts in this direction are multiplying. A big challenge in magnetoplasmonics, is to mitigate the presence of the losses, which hinder the possibility to have large magneto-optical effects. This is true also for 2D systems and in the case of 3D systems, high losses are even more detrimental since they would reduce the penetration of light inside a 3D magnetoplasmonic crystal. Even ferrimagnetic dielectrics are lossy compared to semiconductors and transparent oxides for photonic applications, and this is a limitation in view of practical devices where magnetic and plasmonic functionalities can be exploited. Interestingly, the band structure of 3D crystals might help in this respect since waves with frequencies within the forbidden gap should give rise to very sharp and low loss propagating modes with a great potential for sensing and quantum photonic applications [8]. However, such waves are transported only over a limited distance, the so-called Bragg length, thus limiting the full exploitation of the magneto-optical interactions along the third dimension in magnetoplasmonic crystals. Moreover, the excitation of volume collective modes in 3D plasmonic and magnetoplasmonic crystals is predicted to produce reflective modes resilient to the plasmonic absorption, exhibiting reflection that approaches unity with extremely low loss, as well as the emergence of lattice matched scattered waves leading to full transmission through a spectral transparency window [9]. These attractive optical properties of the volume collective modes may lead to a breakthrough in the design of low-loss and efficient 3D magnetoplasmonic metamaterials for novel linear and nonlinear photonic applications. Thus, sending waves much deeper into these crystals represents both an opportunity and a big challenge. Furthermore, it is worth mentioning that 3D magnetoplasmonics might be key for the advancement of light-driven magnetism. It has been recently shown that plasmonic vortices carrying orbital angular momentum can be generated and manipulated using relatively low magnetic fields [10], and the inverse effect to induce topological spin textures (see Sections 9 and 11)might be achieved in the near future.

Finally, the perhaps biggest challenge is the ability to fabricate and characterize (see also Sections 1, 2, 4, and 5) high quality hybrid 3D magneto-plasmonic structures, as this represents the first necessary step to achieve suitable 3D magnetoplasmonic systems to investigate all the intriguing physics described above.

8.3 Advances in Science and Technology to Meet Challenges

Designing the complex plasmonic structures described above is challenging due to their multiscale nature since computational modelling of nanoscale building blocks require very fine spatial discretization of the computation domain and, in in the case of finite size 3D crystals, also large simulation domains. To tackle this challenge, two approaches are generally taken: (i) the effective medium approximation, which is computationally light but neglect the nanoscale effects and (ii) the use of a unit cell with periodic boundary conditions. The latter approach fails to describe the photonic properties arising from their finite-size. In this context, it has been shown that the finite-difference time-domain method can be used to accurately calculate the photonic band structures of finite crystals at different size scales (Figure 8.2a) [11]. The same methodology, combined with discrete adjoint method, allows to carry out inverse design of plasmonic structures using density-based topology optimization and achieve desired optical properties by predicting a complex structure geometry [12]. Additionally, it was recently shown that data-driven exploration of the possible space of photonic band gap crystal structures is also a promising approach, as it was suggested that many current data-driven heuristic approaches provide general guidance to designing crystals for photonic band gaps. Regarding the Bragg length limitation hindering the penetration of waves with frequencies within a forbidden gap mentioned above, two main strategies can be followed. By spatially shaping the wavefront, the internal energy density can be enhanced at a tuneable distance away from the front surface, up to one order of magnitude more than the Bragg length [8]. At the same time, it is possible to penetrate deeply inside a plasmonic crystal by exploiting the coupling between the surface propagating plasmon modes and the underneath crystal structure [13].

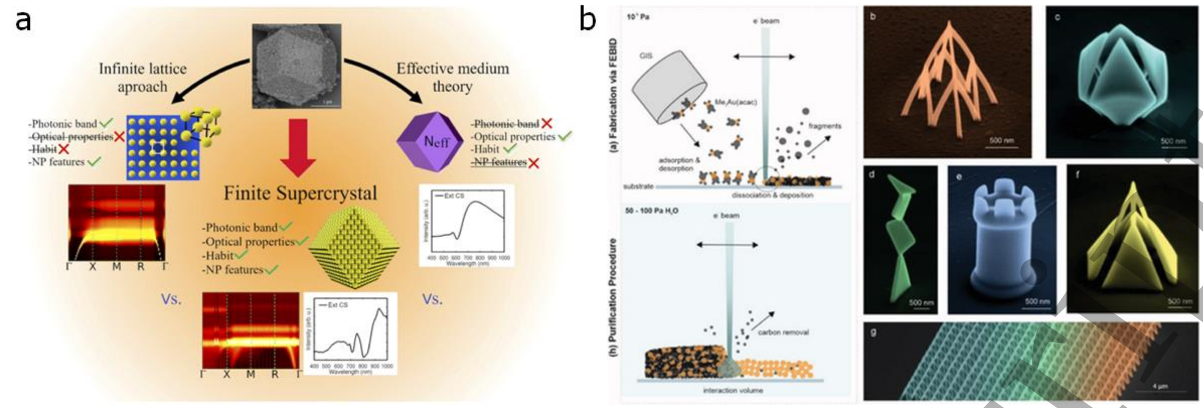

Figure 8.2. (a) Approaches to calculate the properties of photonic crystals, i.e., infinite lattice for band structure calculations, effective medium theory for optical properties like reflectance, and proposed FDTD simulation for finite supercrystals. (b) Fabrication, purification, and 3D possibilities. Top-left side: a gas injection system (GIS) injects preheated precursor molecules in gaseous states (Me2(acac)Au(III) in this study) in close proximity to the substrate, where they adsorb, diffuse and, if not dissociated, eventually desorb again. Upon interaction with a focused electron beam, local dissociation leads to immobilization of the respective metal, which forms the intended deposit. As a result of incomplete ligand cleavage not only Au atoms (orange spheres) but also carbonaceous fragments (grey spheres) are co-deposited, which is why FEBID structures naturally contain high amounts of carbon. The whole fabrication procedure is typically performed at 21 ± 1°C and 10−5 Pa. Right panel: 3D nano-printing design possibilities, ranging from meshed objects over sheet-like, semi-closed toward closed and mixed architectures. Also, scalability is provided, to generate micrometre-sized assemblies for individual applications (see tetrapod array). To remove the carbon, the scheme on the bottom-left panel illustrates a suitable purification process, which takes place in a low-pressure H_2O atmosphere ranging between 50–100 Pa at 21 ± 1°C. Using again the focused electron beam leads to the local removal of carbon, leaving nominally pure, polycrystalline gold nanostructures with a volume loss of up to two thirds. Depending on the height of the nanostructure, the beam conditions must be adapted such that the interaction volume can cover the whole structure volume. Panel (a) is reproduced from Ref. [11]. Panel (b) is taken from Ref. [14].

At the fabrication level, currently there are few techniques that allow 3D-printing of nanoscale hybrid plasmonic and magnetic structures. One example is direct laser writing using multi-photon polymerization to create 3D polymer scaffolds followed by the deposition of metallic layers. The resolution of this approach is limited, although sub-micrometric, and multiple materials, e.g., plasmonic and magnetic, can be deposited only in form of multilayers. The most promising approach is the use of focused electron-beam-induced deposition (FEBID), a highly flexible additive 3D direct-write technology with spatial nano-scale precision (Figure 8.2b). A computer aided design approach supplants trial and error toward more precise/accurate FEBID required for the realization of suitable 3D nanostructures, to achieve customized plasmonic responses, therefore paving the way for yet unrealized plasmonic applications in 3D space [14].

8.4 Concluding Remarks

In summary, we have discussed how magnetoplasmonics in 3D can open new avenues towards a superior control of magneto-optical and opto-magnetic effects stemming from the richer 3D photonic band structure combined with unique 3D spin textures. The full exploitation of these 3D features requires that light can penetrate deep inside the magnetoplasmonic structure. This poses a serious challenge given the lossy nature of materials, typically metals, utilized for implementing such 3D systems. Recent encouraging results have shown that this challenge might be circumvented by proper shaping of light waves and by exploit the coupling between photonic modes along the out-of-plane direction. Further advancements in the theoretical understanding and modelling are therefore required to enable quantitative and reliable predictions for such complex systems. To face such challenges, data-driven approaches combined with inverse design and finite difference time domain methods look promising to design and predict the optical behaviour of complex 3D magnetoplasmonic crystals. The other grand challenge is the precise nanofabrication of such 3D systems. Thanks to the impressive progress in the fabrication of complex multi-material 3D structures with nanometric control, approaches based of focused ion beam milling and focused ion/electron beam induced deposition seem to be the best candidates to achieve the objective.

Acknowledgements
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9- 3D topological spin textures

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9.1 Status

The energetic ground state of a magnetic material is the result of various competing local and non-local magnetic interactions among the individual spins and is characterized by a specific arrangement of all spins in so called spin textures or magnetic domains. The distribution of magnetization vector fields can be associated to mathematical entities and, thus, to the concept of topology which allows to classify geometrical properties of continuous structures. Two structures are considered topologically equivalent if there is a continuous mapping from one to another. In the case they are not equivalent, a continuous transformation is forbidden thereby providing a finite energy barrier in a physical system – known as *topological protection* – between these two vector field distributions [1].

Magnetic skyrmions are among the most prominent examples of topological spin textures that have been recently intensely studied both for fundamental and applied reasons [1]. They are considered as 2D spin textures, characterized by a unitary *skyrmion number* which is defined as $N_{sk} = \frac{1}{4\pi}\iint d^2r\mathbf{m}\cdot(\frac{\partial \mathbf{m}}{\partial x}\times\frac{\partial \mathbf{m}}{\partial y})$, with **m** being the unit vector of the magnetization. Topologically, magnetic spin textures with the same skyrmion number are equivalent.

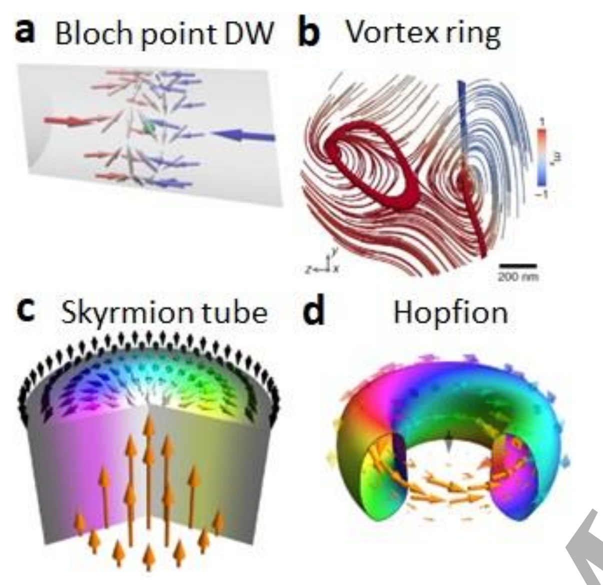

Figure 9.1. Schematic spin textures of (a) a Bloch point type domain wall, reproduced with permission from Ref. [2], (b) vortex ring, reproduced with permission from Ref. [4], (c) a skyrmion tube, and (d) a Hopfion with *Hopf index* = 1, reproduced with permission from Ref. [5].

Whereas this is true for systems, where the thickness of the magnetic material exceeds material specific exchange lengths, the 2D characters of spin-textures evolve in more complex objects with a non negligible 3rd dimension [2] (see Figure 9.1 for four examples). The 3rd dimension changes dramatically the properties and behavior of spin textures. The velocities of domain walls (DWs) in 3D should be much larger than their 2D counterparts. For example, Bloch point type DWs show absence of the Walker breakdown and their velocity exceeds even the speed of sound in 3D nanowires [3]. Magnetic vortices can be observed in the form of a 3D vortex – antivortex pair called a vortex ring [4]. Trivial extensions into the 3rd dimension for skyrmions are skyrmion tubes found both in bulk and multilayer systems [5]. More interesting are non-trivial 3D topological spin textures such as chiral bobbers [5], skyrmionic cocoons [6], [7], or Hopfions [2]. The latter can be classified by an additional topological quantity, i.e. the *Hopf index* [8] $H = \frac{1}{(4\pi)^2}\iiint d^3r\mathbf{F}\cdot\mathbf{A}$, where $F_i = \varepsilon_{ijk}\mathbf{m}\cdot\frac{(\partial_j\mathbf{m}\times\partial_k\mathbf{m})}{2}$. Here, $\varepsilon_{ijk}$is the Levi-Civita tensor and $i,j,k = \{x,y,z\}$. $\mathbf{A}$ is a vector potential such that $\mathbf{F} = \nabla\times\mathbf{A}$. A magnetic Hopfion can be envisioned by twisting a skyrmion tube along the tube axis, followed by connecting the two ends of the twisted tube into a torus. Lines with constant magnetization direction (isolines) circle around the surface of the torus. Under certain conditions, the Hopfions motion does not exhibit a topological Hall effect, which leads to an unwanted sideways motion in magnetic racetracks. Recently, Hopfions have been experimentally observed in bulk [9] and multilayer systems [10].

9.2 Current and Future Challenges

3D topological spin textures are of great interest in current and future research of fundamental phenomena in magnetic materials, but are also key to understand properties and functionalities in future spintronic applications. To advance those areas of research and technology, challenges in the design and theoretical understanding of 3D topological spin textures with bespoke static and dynamics properties need to be overcome (see Figure 9.2).

The utilization of 3D textures in applications will benefit from electrical manipulation. Reliable and efficient protocols for the nucleation of 3D spin textures driven by electrical currents are necessary. While current-driven dynamics have been predicted, there is still a large gap in experimental evidence, except for a first demonstration for magnetic cocoons [7], and skyrmion tubes[11]. Since the electrical signature could depend on the Hopf index, this might open the way to use the Hopf index as a design parameter for future innovative 3D storage devices.

Frustration is commonly used to stabilize 3D textures and frustrated magnets exhibit non-standard contributions to the spin-transfer torque (STT) and topological Hall effect. Modeling, measuring, and analyzing the response of frustrated 3D textures to these novel terms will be important to expand their dynamical features.

Neuromorphic computing requires a precise control and local manipulation of 3D spin textures, which is challenging due to local variations of parameters. Proper field/current protocols that can be applied in a reduced region only need to be developed for this purpose. A recent prediction in neuromorphic computing is the use of Hopfion interactions with spin-waves [12], which would launch a new future research direction, i.e., *Hopfion Magnonics*. Prerequisite will be an accurate analysis to understand how 3D spin textures can interact with spin-waves (scattering, soliton dynamics, deformations), and how 3D textures could generate spin-waves. The latter could serve as a fingerprint for the identification of the 3D spin texture itself.

Future synthesis of 3D topological spin textures benefits from recent developments in advanced nanostructuring techniques, e.g. to overcome the limitation of 2D lithography by 3D laser lithography. Here, two-photon lithography is an emerging tool and has already demonstrated a 3D replica of an artificial spin ice system. The most versatile synthesis tool is probably focused electron beam induced deposition (FEBID), which enables to fabricate with an almost CAD approach very complex multicomponent magnetic structures, such as tetrapods, curved nanowires or bucky-ball structures.

To validate the properties and behavior of 3D topological spin textures, significant advances in characterization are required, specifically to image with a spatial and temporal resolution down to fundamental magnetic length and time scales the full 3D arrangement of the spins.

9.3 Advances in Science and Technology to Meet Challenges. Modelling of 3D topological spin textures needs (i) the development of and sufficient access to large computational facilities to allow the complete computation of magnetostatic interactions being important in the stabilization of these textures, and (ii) highly sophisticated models including higher order exchange interactions. Multi-GPUs micromagnetic solvers can be used to achieve those objectives.

From an experimental point of view, further advances in synthesis and fabrication capabilities of 3D topological spin textures are needed. For example, the integration of Artificial Intelligence/Machine Learning will drive new processes in additive manufacturing, which could benefit 3D nanomagnetic systems. Of equal importance for the synthesis of 3D topological spin textures are recent advances in nanostructuring techniques. An example is the electrochemical deposition to synthesize cylindrical magnetic nanowires via engineered highly ordered (hexagonal lattice) nanoporous anodic aluminum oxide (AAO) templates with controllable diameter and interpore distances, that led e.g. to free-standing low density interconnected metallic nanowire networks.

Advanced characterization of 3D topological spin textures is essential to validate and to detect both spin-waves and topological textures and how they interact. Among the most promising directions, there are current developments in techniques notably using electron and x-ray tomography based on state-of-the-art electron microscopes as well as novel x-ray techniques, such as x-ray laminography and ptychography. Finally, co-design approaches spanning from basic material science to devices, algorithms and systems are worth considering to accelerate a deep integration of spintronic devices with 3D topological spin textures towards their technological applications. Figure 9.2 summarizes the concept of this co-design approach.

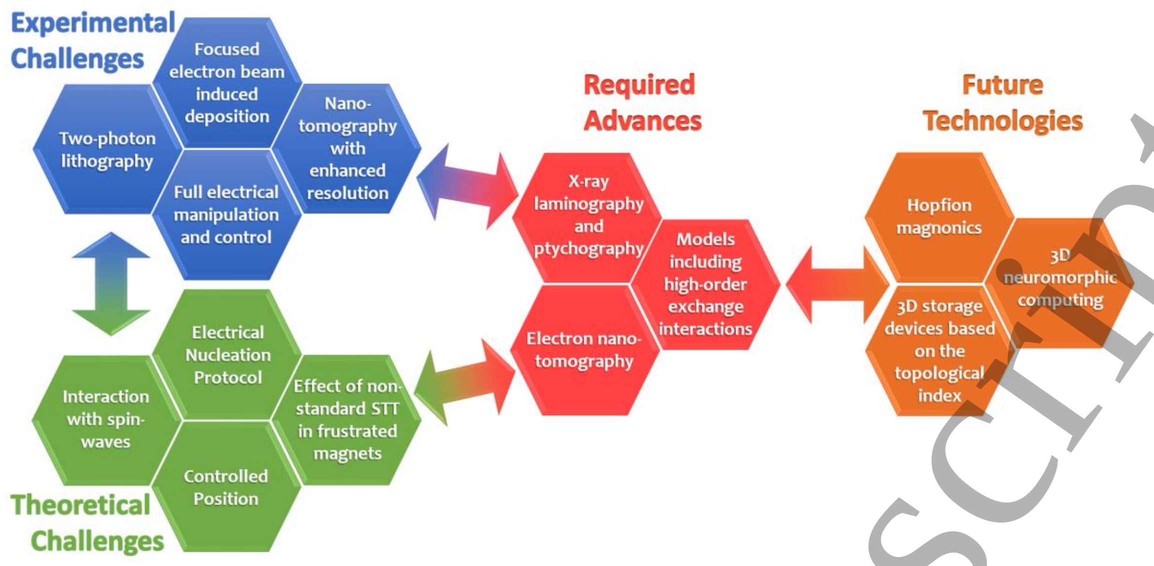

Figure 9.2. Concept of a co-design approach towards futured 3D technologies with spin textures

9.4 Concluding Remarks

The research on topological aspects in 3-dimentional spin textures is a rapidly emerging field of research and has already created a wealth of new research directions with novel concepts that are highly relevant for future technological applications. Non-trivial 3D topological spin textures, which might be advantageous compared to 2D skyrmion systems are so far much less investigated due to the challenges as discussed here. The achievement of new theoretical and experimental tools will have the potential to develop new building blocks for future technologies (see Figure 9.2), and meet technological challenges related to high density, 3D interconnection and ultralow power neuromorphic computing, as well as low power microelectronics in general. A very promising route for *Hopfion Magnonics* seems to have opened recently[12]. To conclude, extending 2D skyrmion systems into the 3rd dimension will enable systems with higher complexity and leverage chirality as an inherent 3D phenomenon that has impacted nature from the smallest to the largest length scales.

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10 – Molecular Magnets

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10.1 Status

Molecules have played a key role in nanomagnetism, particularly in evidencing quantum effects in macroscopic magnetization.[1] The tuneability of the magnetic parameters, the quantum nature of the molecular spin, the strict monodispersed character in ensemble measurement, and the ease of processing, which has made single-molecule magnetic investigations accessible, are the cornerstones on which molecular nanomagnetism has been flourishing in the last decades.

Magnetic anisotropy has been the critical parameter to optimize to induce a large potential barrier for the reversal of the magnetization of individual molecules, called single-molecule magnets.[1] These magnetic molecules became thus a model of zero-dimensional magnetism. Spectacular progress has been made in the chemical design of highly anisotropic systems, though at the expense of the chemical robustness of the molecular scaffold.[2] Indeed, metal ions with a low coordination number – thus more reactive – favour sizeable magnetic anisotropy. If, on the one hand, miniaturization has processed down to evidencing magnetic bistability in a single atom deposited on a non-magnetic oxide, on the other, the operative temperatures remain far from room temperature. This is not a significant drawback for quantum applications that are still restricted at cryogenic temperatures.[3] Most quantum applications necessitate long coherence of the superposition of quantum states, which in electron spin systems is optimized when the spin-orbit coupling is minimized, and the environment is nuclear spin-free. Achieving the latter condition is particularly challenging in molecular materials, but this disadvantage is effectively countered by the high tunability of the spin Hamiltonian at the synthetic level, thereby expanding the arsenal of quantum logic tools available.[4]

Control of magnetic exchange interactions between the molecular units is also mandatory to harness the vast potential of molecular chemistry in magnetism. Here, the chemical design has benefited from the qualitative but very helpful Goodenough – Kanamori rules established in the ’50 based on the symmetry of the orbitals carrying the unpaired electron or involved in the super-exchange pathways. Quantum chemistry has also contributed significantly, and DFT calculations can predict exchange coupling constants ranging from thousands to hundredths of kelvin with great accuracy. When assembling magnetic units into one-, two- or three dimensions, the architects of the matter can now control both magnetic anisotropy and magnetic interactions concurrently with the spatial arrangement of their building units. This control of matter at the molecular level has given access, for example, to unprecedented magnetic behaviours like single-chain magnetism[5] and has enabled the design of unprecedented lightweight magnets as the one shown in Figure

10.1 [6, 7] that order far above room temperatures and possess a coercivity that compares well with the inorganic magnets used in our modern life.

Modern coordination chemistry and molecular chemistry offer to the field of molecular magnetism a quasi-infinity of opportunities to design at will the next generation of magnetic materials with premediate and controlled physical properties.

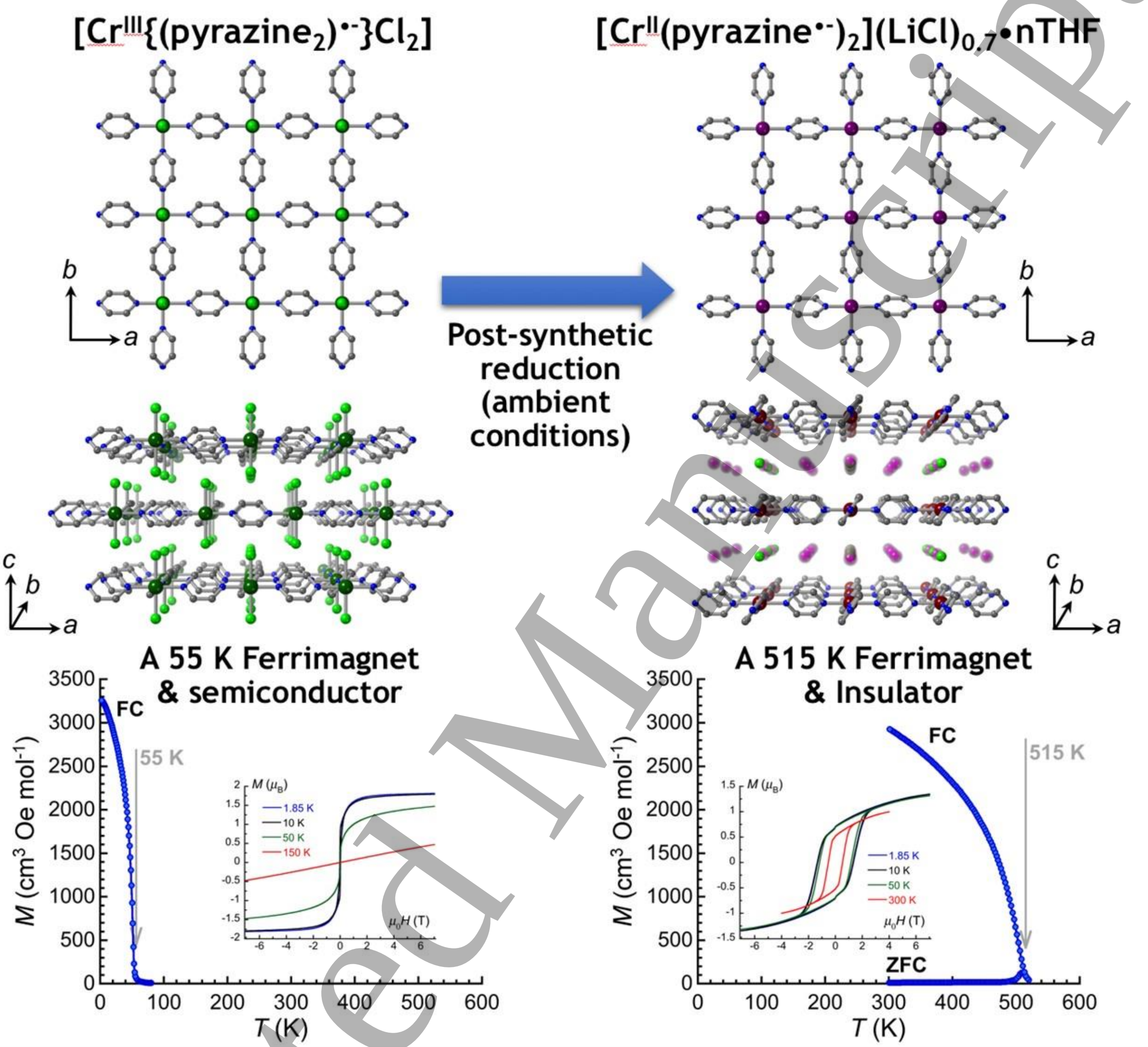

Figure 10.1. Scheme illustrating the post-synthetic reduction of the pre-formed $[Cr^{III}\{(pyrazine_2)^{\bullet-}\}Cl_2]$ pyrazine-based coordination network to form the 515 K ferrimagnet, $[Cr^{II}(pyrazine^{\bullet-})_2](LiCl)_{0.7}\bullet nTHF$. Their associated magnetic properties (as *M* vs *T* and *M* vs *H* plots) are also shown on the lower part of the figure. Colour code: C, grey; N, blue; Cr^{III}, dark green; Cr^{II}, dark purple; Cl, light green; Li, purple. Adapted from references [6, 7].

10.2 Current and Future Challenges

One of the key issues in molecular magnetism, but more generally in magnetism, is the simultaneous maximization of magnetic exchange interactions and magnetic anisotropy. The first requires significant electronic delocalization, while the second involves retaining unquenched orbital contributions to the magnetic moment. The latter are maximized in three positive lanthanide ions where the unpaired electrons reside in the well-shielded 4f orbitals, which do not promote significant orbital overlap and strong magnetic exchange. Many efforts have been devoted to the use of bridging ligands carrying unpaired electrons, with the $N_2^{3-\bullet}$ radical having the highest efficiency in transmitting the exchange interaction. Unfortunately, the obtained materials are only stable at low temperatures and in an inert atmosphere.

Learning from traditional magnetism, mixed valence in oxides promotes efficient ferromagnetic interactions through the 'double exchange' magnetism. The unprecedented realization of a mixed-valence $Gd^{II/III}$ organometallic dimer, whose structure is shown in Figure 10.2, has demonstrated that the exchange interaction can be as large as 1000 K, overwhelming any previous report. Anisotropic lanthanides such as dysprosium in the isostructural molecule exhibit a coercive field larger than 14 T for temperatures below 60 K.[8] These disruptive results immediately pose the question of whether a chemical approach based on mixed-valence in lanthanides can be developed to achieve an extended and chemically robust lattice for ultra-hard magnets like it was done recently for 3d metal ions such as chromium.[6, 7] Combining the mixed valency of metal ions and/or organic bridging ligands with a proper orbital synergy into two- or three-dimensional networks appears to be a strategy of choice to promote long-range ferro- or ferri-magnetic order at ambient temperature in molecule-based materials. These future systems and their large electron and spin delocalization on extended networks also offer the opportunity to engineer materials with high conductivity playing on the stoichiometry of the electrons delocalized on the network.[9]

Molecular magnets also have a great potential in nanomagnetism for the possibility to exploit supramolecular, i.e. non-covalent interactions such as van-der-Waals, π-stacking, or hydrogen bond interactions, to obtain magnetic nanoarchitectures from soft bottom-up and self-assembly approaches. Magnetic exchange mediated by van-der-Waals interactions is currently investigated in 2D or layered inorganic lattices but remains unexplored in molecule-based materials. A recent investigation has shown that magnetic interactions up to ca 150 K can be established between Cu^{II} spins coordinated by sulfur-rich ligands through an intermolecular S···S pathway.[10] Interestingly, this class of neutral molecules can be evaporated, and thus, the magnetic exchange network is transferrable from the bulk to a substrate and hybrid nano-architectures.

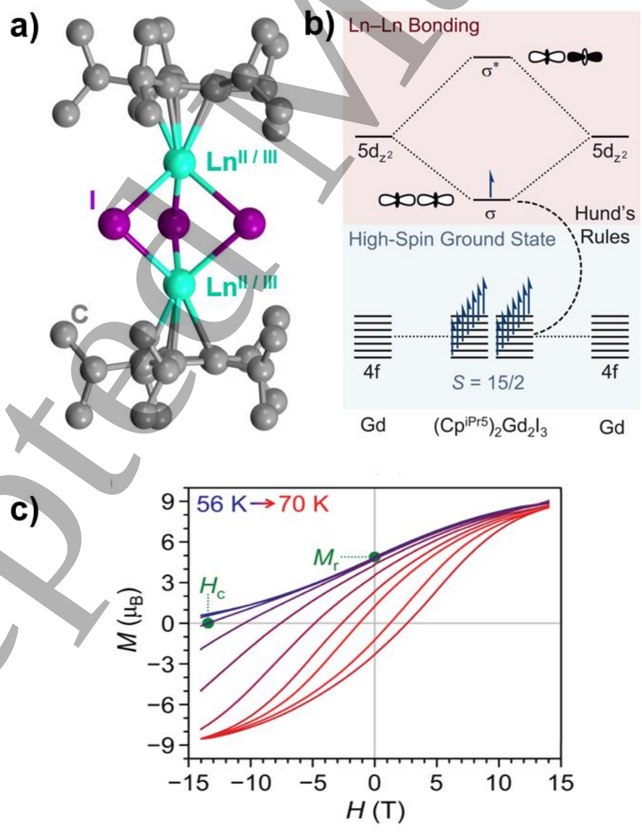

Figure 10.2. a) Structure of the complexes of formula $(Cp^{iPr5})_2Ln_2I_3$ (Cp^{iPr5}= pentaisopropylcyclopentadienyl; Ln = Y, Gd, Tb, or Dy), b) Molecular orbital diagram for $(Cp^{iPr5})_2Gd_2I_3$ illustrating the mechanism leading to strong ferromagnetic exchange interaction through the formation of a singly occupied σ-bonding orbital. c) Temperature dependence of the magnetic hysteresis of the $(Cp^{iPr5})_2Dy_2I_3$ single-molecule magnet showing

extremely hard magnetism. Reproduced from C. A. Gould *et al*, *Science* 375,198 (2022) with permission from AAAS.

10.3 Advances in Science and Technology to Meet Challenges

Unleashing the full potential of molecular nanomagnets for advanced quantum applications requires overcoming the roadblock of single-spin addressing. While the nanosize confinement is easily achieved thanks to the bottom-up approach, the control and readout at the single spin remain an issue. The poor spatial confinement of magnetic fields has been solved by placing molecules in nanojunctions and using transport measurements.[3] The astonishing progress in scanning probe spectroscopies based on spin-polarized tips and microwave excitations has entirely revolutionized the field of nanomagnetism. The accurate control of the position of three magnetic titanium adatoms on an oxide surface has recently allowed the building of a quantum CNOT gate. Interestingly, the magnetic field is controlled at the subnanometric scale thanks to the precise positioning of a neighboring iron atom.[11]

Despite the remarkable achievements obtained with this approach, a pathway for scalability is challenging to devise. Adding an optical interface to molecular nanomagnets[12] seems a viable route toward single molecular spin readout, analogous to what is already achieved with nitrogen-vacancy defects in diamonds. An additional feature that can be implemented in molecular materials in a more efficient way than in inorganic lattices is structural chirality. Combined with magnetism, this can give rise to exotic properties, like magnetochiral dichroism, *i.e.,* the different absorption of unpolarized light depending on the mutual orientation of the magnetic field and chirality vector. More relevant for single-spin addressing in molecular nanomagnets is the phenomenon of chirality-induced spin selectivity. Many reports have demonstrated that chiral media act as spin filters in electron transport at room temperature. More recently, the phenomenon has been observed also in light-induced electron transfer at the molecular scale. The interplay between the spin state and the transfer rate of electrons to form charge-separate states or to give charge recombination opens the way to an innovative spin-to-charge conversion facilitating the spin readout. This could significantly boost the potential of molecular materials for quantum applications.[4, 13]

10.4 Concluding Remarks

Molecules represent a still underexplored resource for magnetism, particularly nanomagnetism. The chemical design can control several key features, ranging from magnetic anisotropy to exchange interaction, thus resulting in high coercivity. The intrinsic confinement at the nanoscale and the easy processability make magnetic molecules an ideal playground for the development of spin-based quantum technologies. Drawbacks are also present. Best-performing molecular systems often lack the high chemical stability necessary for technologically relevant applications. In quantum nanoscience, magnetic molecules have unparalleled potential, but developing efficient tools for single-spin readout in molecular materials is the big challenge we have to face.

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11 - Curvilinear magnetism

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11.1 Status

The research field in magnetism dealing with the impact of geometric curvature on magnetic responses of curved 1D wires and 2D shells is known as curvilinear magnetism. The expression "curvilinear magnetism" was introduced back in 2016 by Streubel *et al* [1] and is currently used to describe consolidated activities of the fundamental and application oriented interdisciplinary community of physicists, chemists, material scientists and biologists [2]. In addition to device ideas in memory and logics (see Sect. 17), curvilinear magnetic architectures are considered for brain-inspired computing and 3D interconnectivity and enable novel applications scenarios including artificial fertilization, skin conformal and printed magnetoelectronics. The latter explorations already resulted in technology transfer activities aiming to commercialise mechanically re-shapeable spintronic devices. (see Sect. 18)

In this roadmap, we deliberately focus on prospective fundamental developments in the field of curvilinear magnetism.

Over about a decade of an intensive research, curvilinear magnetism emerged from a neat mathematical abstraction to a powerful micromagnetic framework, which is used to predict new physical phenomena and guide experimental research. This is a mathematically strict theory which resembles a new leap in the contemporary micromagnetism. Technically speaking, within the curvilinear magnetism approach, the geometric properties of a magnetic object are embedded in the micromagnetic energy functional to explain experimental observations or results of micromagnetic simulations (see Sects. 13 and 16) in terms of effective anisotropic and chiral interactions. These interactions emerge from any spatial derivative in the energy functional (exchange, Dzyaloshinskii-Moriya interaction (DMI), magnetostatics, spin transfer and spin-orbit torques) when performing coordinate transformation from the Cartesian to a curvilinear reference frame.

The curvature has influence on the ground state and on the magnetisation dynamics (spin waves, domain walls and skyrmions) in a geometrically curved object.

It is established that magnetochiral responses of any curvilinear ferromagnetic nanosystem are governed by the mesoscale DMI, which is determined via both the material and geometric parameters. Its strength and orientation can be tailored by properly choosing the geometry, which should enable stabilizing distinct magnetic chiral textures including skyrmion and skyrmionium states as well as skyrmion lattices. In 2020, the local theory of curvilinear magnetism initially proposed by Gaididei *et al* [3], was extended to account for nonlocal interactions like magnetostatics [4]. This curvilinear micromagnetic framework allows to predict existence of a novel nonlocal chiral symmetry breaking effect, which is responsible for the coexistence and coupling of multiple magnetochiral properties within the same curvilinear magnetic nanoshell [5].

The relevance of the curvilinear magnetism is dictated by the active exploration of geometrically curved magnetic architectures in fundamental research and technology. (see Sects. 1 and 2) The impact of geometric curvature on magnetic textures in curved shells is categorised in a convenient textbook manner [4] to ease its utilisation in fundamental and application-oriented research, which currently affects each and every topic in modern nano-magnetism including spintronics, (see Sect. 18) spin-orbitronics and magnonics (Figure 11.1). (see Sects. 14 and 15)

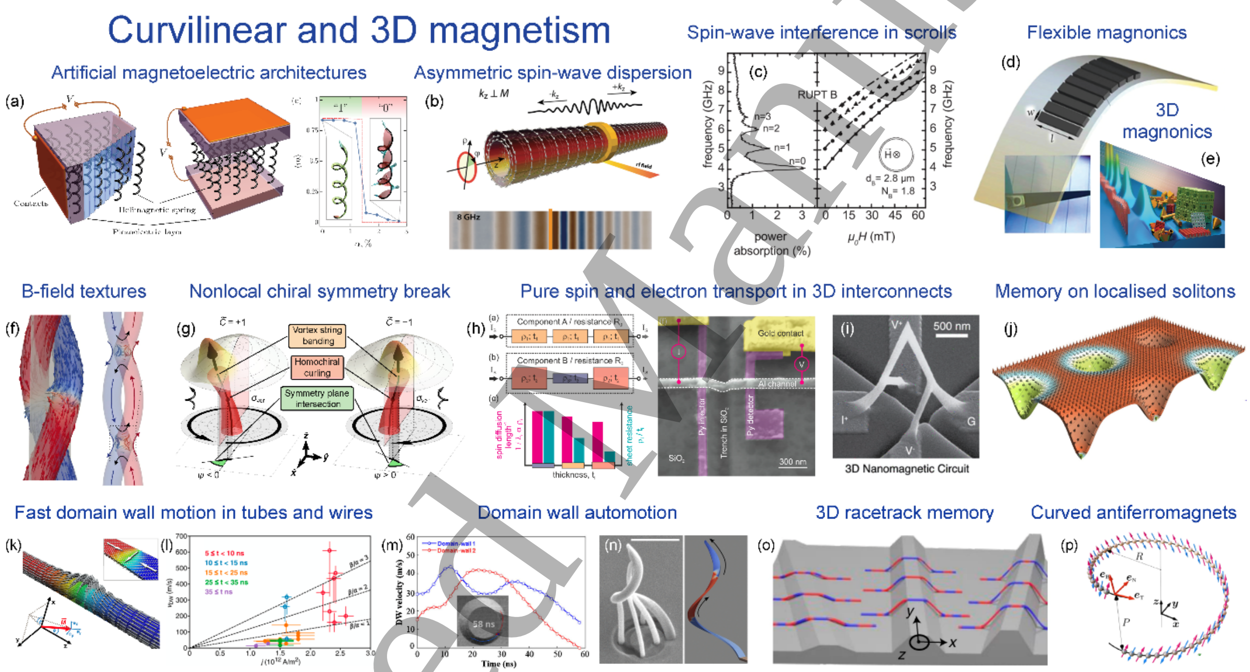

Figure 11.1: Research directions within the curvilinear and 3D magnetism. All panels are reproduced with permission from the following papers: (a) Volkov OM *et al* 2019 *J. Phys. D: Appl. Phys.* **52** 345001; (b) Otálora JA *et al* 2016 *Phys. Rev. Lett.* **117** 227203; (c) Balhorn F *et al Phys. Rev. Lett.* **104** 037205; (d) Reprinted from Faurie D *et al* 2021 *J. of Appl. Phys.* **130** 150901; (e) Gubbiotti, ed.: Three-Dimensional Magnonics: Layered, Micro- and Nanostructures. Jenny Stanford Publishing (2019); (f) Donnelly C *et al* 2022 *Nat. Nanotech.* **17** 136; (g) Ref. [5]; (h) Das KS *et al* 2021 *Nano Lett.* **19** 6839; (i) Meng F *et al* 2021 *ACS Nano* **15** 6765 (2021); (j) Kravchuk V P *et al* 2018 *Phys. Rev. Lett.* **120** 067201; (k) Yan M *et al* 2010 *Phys. Rev. Lett.* **104** 057201; (l) Schöbitz M *et al* 2019 Phys. Rev. Lett. **123** 217201; (m) Mawass M-A *et al* 2017 *Phys. Rev. Appl.* **7** 044009; (n) Skoric L *et al 2022 ACS Nano* **16** 8860; (o) Gu K *et al* 2022 *Nat. Nanotech.* **17** 1065; (p) Pylypovskyi O V *et al* 2020 *Nano Lett.* **20** 8157.

11.2 Current and Future Challenges

Magnetomechanics: the current theory of the curvilinear magnetism does not address the effects of strain (or in general magnetomechanics). This makes the current theory incomplete and does not allow to have a complete understanding of the processes, which happen for instance in flexible magnetic sensor devices upon mechanical deformation. Therefore, one of the important milestones in the development of the theory of curvilinear magnetism is its extension to tackle the effect of strain in curved architectures, which is relevant for any practical assessment of the performance of devices based on these low-dimensional systems.

Magnetic ordering beyond ferromagnetism: Curvature effects are considered primarily in ferromagnetic systems. Considering their relevance for energy efficient and ultrafast spintronics, it is insightful to extend the theory to antiferromagnets. Fundamentally, they offer a boarder range of coupled magnetic order parameters and interactions including weak ferromagnetism and homogeneous DMI. Curvilinear antiferromagnets offer geometrically tunable orientation of the Neel vector, the helimagnetic phase transition and hybridization of spin wave modes [6]. The perspective of these predicted curvature effects for engineering of chiral and anisotropic responses and soliton dynamics for applications is not explored by now.

Magnetoelectric responses in curvilinear architectures: Recently, Ortix *et al* made a theoretical proposal towards realising magnetoelectric responses relying on zig-zag shaped ferromagnetic nanowires with DMI featuring alternating magnetization [7]. The experimental activities on this appealing topic, which makes a link towards multiferroic community, are not started yet.

From micromagnetic to B-field textures: Recently, Donnelly *et al* went beyond the consideration of micromagnetic textures and addressed magnetic field nanotextures peculiar for 3D curved nanowires [8]. The conceptual novelty is the demonstrated possibility to realise textures in the magnetic induction like cross-tie B-field textures. (see Sect. 5) In contrast to the topological magnetisation textures like domain walls and skyrmions whose technological relevance is already established, the application potential of B-field nanotextures is yet to be understood and explored.

Effects of topology (see Sect. 9): Curvilinear magnetism couples topology of the magnetic texture and topology of the geometric shape of the object. This constrains the properties of the magnetic vector fields in curvilinear magnetic architectures. For instance, it can be anticipated that vorticity of magnetic surface textures, like surface vortices and antivortices living on curvilinear shells represented by minimal surfaces, is determined by the Euler characteristic of the object. This can provide appealing possibilities to fabricate objects supporting higher order vorticity states accommodating virtually infinite number of textures per object in the ground state as needed for reservoir computing or formation of complex magnetic near fields.

11.3 Advances in Science and Technology to Meet Challenges

It is insightful to involve cross-scale methods to realize curvilinear architectures from atomic to mesoscale. Many fabrication methods e.g. based on scanning tunnelling microscopy manipulation, DNA-origami, and use of novel magnetic 2D materials bear strong potential for the curvilinear magnetism community but they are not yet explored. The typically applied methods like focused electron or ion beam induced deposition, (see Sect. 2) strain engineering origami-based approaches, two photon laser lithography (see Sect. 1) should be developed further to extend the portfolio of available curvilinear structures towards not only ferro- but also ferri- and antiferromagnetic nano- and microstructures as well as exchange coupled composites. (see Sect. 6) The availability of curved magnetic architectures motivates development of characterization tools based on x-ray (including magnetic laminography and ptychography) and electron tomography and holography methods. (see Sect. 4) Regarding the x-ray methods, the breakthrough is expected with the next generation synchrotron sources enabling time efficient tomographic reconstructions of the magnetic states in curved objects. As currently both magnetization textures and field textures are becoming of relevance, exploring the full potential of curvilinear magnetic architectures requires complementary techniques, which provide access to the magnetization texture (say, x-ray-based imaging) and to the corresponding field textures. (see Sect. 5) For the later, not only electron-based imaging but also magnetic force microscopy as well as Nitrogen vacancy center scanning magnetometry could be of relevance. (see Sects. 4 and 7)

The activities on strain effects require high quality 2D and 3D curved magnetic structures. In this regard, Fe-filled carbon nanotubes provide access to high quality crystalline Fe nanowires with parameters close to those of bulk Fe. Mechanical deformation of these wires can offer a possibility to deterministically tailor strain distribution in a curved wire. These samples could enable new fundamental insights in the physics of curvilinear magnetism with magnetostriction-related effects.

Considering their broad experimental availability, magnetic wireframes like N-pods and N-torus of soft magnetic materials are expected to gain major interest due to the possibility to stabilize magnetic textures of higher order vorticity. These objects are of relevance for magnonics and unconventional computing. Relevant experiments on the realization of physical magnetic reservoirs are pending as the integration of curved architectures in electronics circuits should be established. Furthermore, magnetic wireframes can be used to design topological magnetic field nanotextures offering vast capabilities in shaping of the magnetic near fields as relevant superconducting electronics, e.g. for pinning of superconducting vortices aiming to control the electrical resistance of superconductors [9,10]. The potential of curvilinear magnetic architectures for “magnetricity”, which is concerned with the steering of emerging magnetic monopoles in 3D artificial spin-ice systems starts to gain attention as well.

11.4 Concluding Remarks

Conceptually, curvilinear magnetism offers a new approach to material science. Indeed, we can predict how any conventional magnetic film (say, Co or Permalloy) should be geometrically curved to enable anisotropic or chiral responses, which are needed for the specific application scenario. Such a geometric tailoring of magnetic responses is complementary to the usual materials screening approach where material responses are adjusted by the proper choice of the magnetic material, doping or interface engineering with needed symmetry and needed strength of micromagnetic parameters.

The current curvilinear magnetism is a truly multifaceted research field, which contributes to nanomagnetism, spintronics and spin-orbitronics, curvilinear magnonics, 3D magnonics, soft robotics, biology and medicine, and flexible and printed electronics. Attention has been brought to various application proposals that arise from curvilinear geometries and are at different levels of readiness for technological implementation. Most of them require further deepening of theoretical framework and wait for experimental realization in the form of device prototypes. At the same time, there are already mature technologies, especially those related to shapeable magnetoelectronics, that reached the sufficiently high technological readiness level to anticipate rapid industrial implementation of flexible and printable sensor technologies. We expect that this perspective will stimulate further developments in curvilinear magnetism, their interconnections with other research communities addressing curvature-induced effects in live science, soft and condensed matter, as well as, industrial explorations by high-tech spin-offs and R&D-oriented companies.

Acknowledgements
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12 - Analytical theories for 3D magnetic nanostructures

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12.1 Status

Ultrathin magnetic films have been the subject of extensive research, however, there remains a critical need for a comprehensive understanding of the transition between their 2D and 3D properties. When the thickness d of a magnetic film exceeds a few tens of nanometers, the magnetization $\mathbf{M}$ can describe some texture along the thickness, and even at saturation, spin waves (SWs) show dynamic modifications. Several analytical approaches have been used to describe SW dynamics in ferromagnets. The first is applicable for samples with ellipsoidal shape, which assumes a saturated state and a homogeneous internal magnetic field. It has been used to derive the SW dispersion in thin films with the semi-analytical approach of Damon-Eshbach or Wolfram-De Wames and the analytical dispersion relation of Kalinikos and Slavin [1], particularly useful for thin films, but also for shapes where simple quantization of the wavenumber can be introduced. A second approach assumes a static magnetization ansatz, e.g., 1D domain wall, 2D skyrmion, or a 3D hopfion texture, where the SW spectrum can be obtained after linearisation of the Landau-Lifshitz equation. These models are restricted to a soliton object, and separate approaches are used for soliton dynamics (Thiele equation) and SW dynamics. When nonlinear effects are involved, the amplitude formulation of the interacting waves is extensively used, particularly for describing multimagnon processes [2]. Overall, SW dynamics assume plane-wave solutions with a further extension made by considering the lattice of interacting magnetic elements or perforated magnetic films, where Bloch theorem was used to calculate the SW dynamics of 3D magnonic crystals [3]. The dynamic matrix method (DMM) [4] has also been used to study thick films and magnetization-graded systems [5]. It is a micromagnetic approach that involves dividing the magnetic medium into small elements with known analytical solutions [6].

The complexity of 3D structures introduces a level of intricacy beyond that encountered in the previous models, making the computation of the dynamics very demanding and hardly amenable to analytical or semi-analytical approaches. Inhomogeneous magnetization texture or pattern modifies the potential landscape at equilibrium for spin dynamics and induces a local magnetic field inhomogeneity, additionally to the outer edges of the sample. Developing analytical models is necessary to deepen the knowledge of the physics, to understand the process responsible for the observed experimental or simulated phenomenon, and to support the development of numerical models that will provide the tools for optimization and thus technological exploitation of 3D nanomagnets.

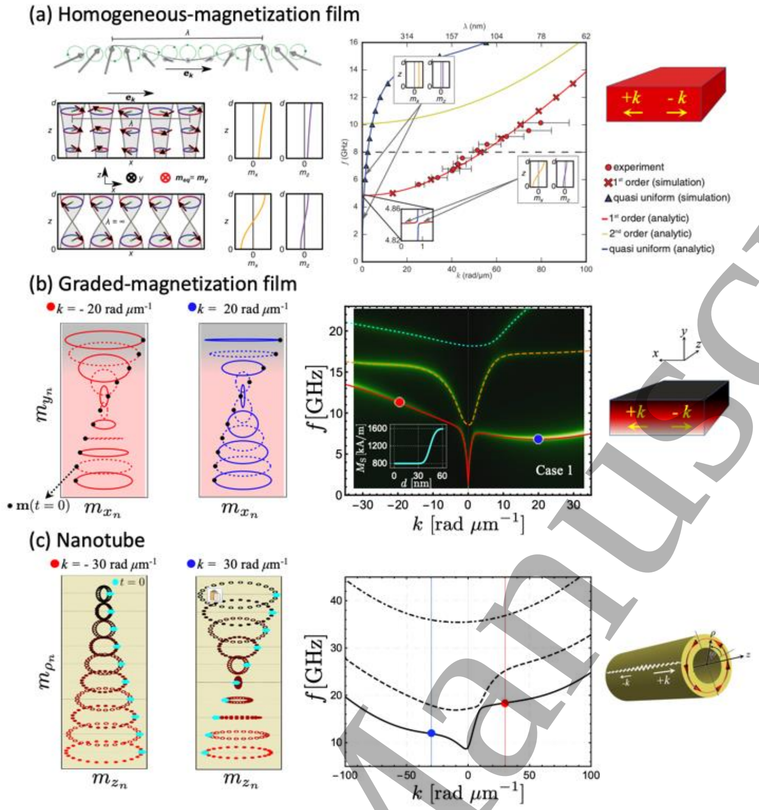

Figure 12.1. (b-c) Spin-wave dispersion and magnetization profiles across the thickness of (a) homogeneous-magnetized film, (b) graded-magnetized film, and (c) nanotube. In (b), the equilibrium magnetization is in plane, while in (c) a vortex ground state is assumed. In all cases a breakdown of the usual quantization condition is observed. Adapted with permission from (a) Ref. [7] (b) Ref. [5] and (c) Ref. [8].

12.2 Current and Future Challenges

Kalinikos and Slavin [Kalinikos86] developed a perturbation SW theory for the case of mixed boundary conditions. In the limit $kd \ll 1$ gives the dipole-exchange spectra for SWs, which can propagate with a wavevector $\vec{k} = \vec{k}_{\parallel} + (n\pi/d)\hat{z}$, being $\vec{k}_{\parallel}$ the in-plane wavevector and n an integer mode number related to the perpendicular standing SWs (PSSWs). The exchange coupling dominates when d is comparable to the exchange length, and the homogeneous mode ($n = 0$) is the low-frequency one. The higher-order PSSW modes ($n \geq 1$) arise when the thickness increases since their frequency is reduced. Nonetheless, the mode hybridization induced by the dipolar coupling destroys the symmetry along the thickness, provoking a shift of the quantized nodal points, as predicted from the off-diagonal elements of the Kalinikos approach [1,7] and the dynamic matrix method [5,6]. In addition, if the system has a magnetic parameter that varies along the normal direction, it is a challenge to derive the correct dispersion relation and quantization of the SW modes.

Recent technological advances enable the fabrication of magnetic graduated nanostructures coupled with different non-magnetic materials and hosting different spin textures depending on the thickness and material features. It opens a realm of possibilities for tailoring the properties and potential functionalities of nanostructures. While graded structures exhibit enriched dynamic properties, they simultaneously elevate the complexity of analytically describing the intricate physics that governs their behaviour. Therefore, the extension of analytical methods, such as the dynamic matrix method, to new geometries that allow non-uniform states is an exciting challenge. Here, the correct treatment of boundary conditions and edge effects is of paramount importance. Indeed, it can lead to boundary-driven twist states (see Fig. 12.2) in the presence of Dzyaloshinskii-Moriya interaction (DMI) [9,10,11]. For instance, free boundary conditions change the orientation of the pitch vector of typical helical states in a bulk DMI films [9], as shown in Fig. 12.2(b-c). Hals and Everschor-Sitte used the generalized DMI to develop the complete form of the boundary conditions at the surface, which reads $2J_{ij}n_i\partial\mathbf{m} = -\mathbf{m}\times(\mathbf{\Gamma}_\mathrm{D}\times\mathbf{m})$, where J_{ij} is a symmetric matrix defining the spin stiffness, $\mathbf{n}$ is the surface normal, $\mathbf{m}$ the unitary magnetization, and $(\mathbf{\Gamma}_\mathrm{D})_k = m_i n_j D_{ijk}$ is the boundary-induced DMI field, which depends on the complete tensorial structure of the DMI tensor D_{ijk}, and not only the antisymmetric part [Hals17]. For thick films with intrinsic DMI, a non-zero symmetric component of D_{ijk} near the interface with a heavy metal [Mulkers18] locally changes the radius of a skyrmion (Fig. 12.2a).

12.3 Advances in Science and Technology to Meet Challenges

Advances in computation technologies have made possible the realistic numerical evaluation of the dipolar coupling in nanostructures, which is the most neglected energy term and expensive in calculation time due to its non-local character. Nowadays, it is known that the local approximation of the dipolar coupling, valid in the ultrathin limit, does not capture its non-local nature (see Sec. 12 on Curvilinear Magnetism), which may cause chiral DW motion and non-reciprocal SW propagation [8,13]. To realistically model 3D nanomagnetism, analytical theories must consider the dipolar coupling, allowing the magnetization to vary across the volume. In the macrospin model, for instance, it is assumed that each nanoelement is homogeneously magnetized and expresses only homogeneous precession in the element. The interaction between the nanoelements is limited to the dipolar and is considered when calculating the ground magnetization state and harmonic oscillations around this equilibrium. This approach become very useful for analysing artificial spin-ice (ASIs), i.e., the lattice of the ferromagnetic nanoelements interacting only by the magnetostatic fields. Recently, the computer code Gænice has been made public [12], which uses this approach for the calculation of SW dynamics in ASIs while allowing for additional magnetization tilt at the nanoelement edges and additional periodic boundary conditions for determination of the SW dispersion in planar structures. In principle, it can be progressed by extending to multilayered nanoelements in planar structures and to 3D lattices.

The DMM represents a powerful tool concerning the dipolar interaction, which relies on the subdivision of the nanostructure into several elements, fully considering the intralayer and interlayer dipolar and exchange couplings and allowing to obtain the matrix elements for the computation of the magnonic spectra. It has been applied to study the SW spectrum of thick nanotubes [3,8], materials with graded magnetization [5], and planar and curved magnetic bilayers [14] (see Fig. 12.1). The DMM can also be combined with the plane-wave method (PWM), which has already been applied to 3D magnonic crystals [3], enabling, for instance, the study of graduated magnonic crystals. Although these methods solve the problem numerically, its matrix elements are described analytically, allowing a better comprehension of the underlying physics. Calculations of the SW dispersion and magnetization profiles across the thickness of homogeneous, graded, and curved films revealed that the distribution of the modes across the thickness is not uniform as previously assumed for the PSSW modes, and a breakdown of the quantization condition occurs (see the Fig. 12.1).

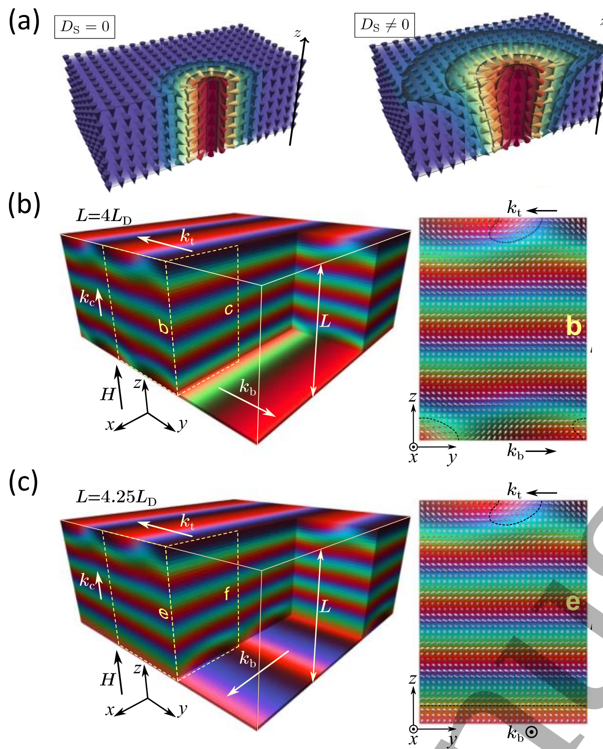

Figure 12.2. (a) Magnetic edge surface states induced by the correct boundary conditions in chiral thin semi-infinite films with bulk DMI [11]. By including the symmetric elements of the DM tensor in the boundary conditions, additional terms appear, increasing the skyrmion radius near the surface. (b-c) Bulk DMI in thick films with free boundary conditions and distinctive surface magnetic states [9]. In the bulk there is a helical state along the field, while at the top and bottom surfaces, the direction of the helix changes depending on the film thickness. Adapted with permission from (a) Ref. [11] and (b-c) Ref. [9].

12.4 Concluding Remarks

As research on 3D magnetic systems evolves, and there is a continuous development of analytical and semi-analytical models that allow for describing and explaining the physical phenomena of SW dynamics observed experimentally or in numerical simulations occurring in these systems. The models originating from thin films and 2D networks of uniformly magnetized nanoelements have already been extended with magnetic inhomogeneity in the direction perpendicular to the plane of the system, such as a dynamical matrix method and an extended macrospin approximation. We can expect soon further development of these models and analytical methods to fully 3D nanostructures with interfacial properties properly considered. It is also expected that other analytical methods known in 2D magnonic systems will be updated to 3D nanostructures, and new concepts known in other areas of physics will be transferred to magnonics. These include the scattering matrix method and Galerkin methods based on expansions in different functional bases or work on the theory of effective parameters, which may allow to describe the SW dynamics at long wavelengths compared to the microstructure dimension of the 3D structure. Developed models will allow to understand the physics of SW dynamics in complex structures and provide the tools for technological exploitation, including neuromorphic computing systems based on 3D magnetic nanostructures.

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13- Analytical and numerical methods - Micromagnetism in 3D nanomagnets

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13.1 Status

Recent progress in magnetic nanofabrication has opened the possibility of investigating three-dimensional magnetic nanostructures, thereby spawning a novel field of research that may pave the way toward new physical properties and functionalities in nanomagnets. By accessing and leveraging unexplored effects occurring only in three-dimensional geometries, three-dimensional nanomagnetism marks a significant departure from the traditional investigation of patterned magnetic thin-film elements as it was conducted intensively over the past decades. Various three-dimensional nanomagnets have recently been investigated in this context, including objects with curved surfaces like nanotubes [1], spherical shells, gyroid structures, or interconnected nanowire arrays [2,3,4]. Studying the magnetic properties in such complex geometries poses challenges for experiment and simulation, necessitating dedicated methods beyond those used in traditional thin-film nanomagnetism. Efficient and reliable micromagnetic simulations are an essential prerequisite for understanding and predicting three-dimensional nanomagnets' physical properties.

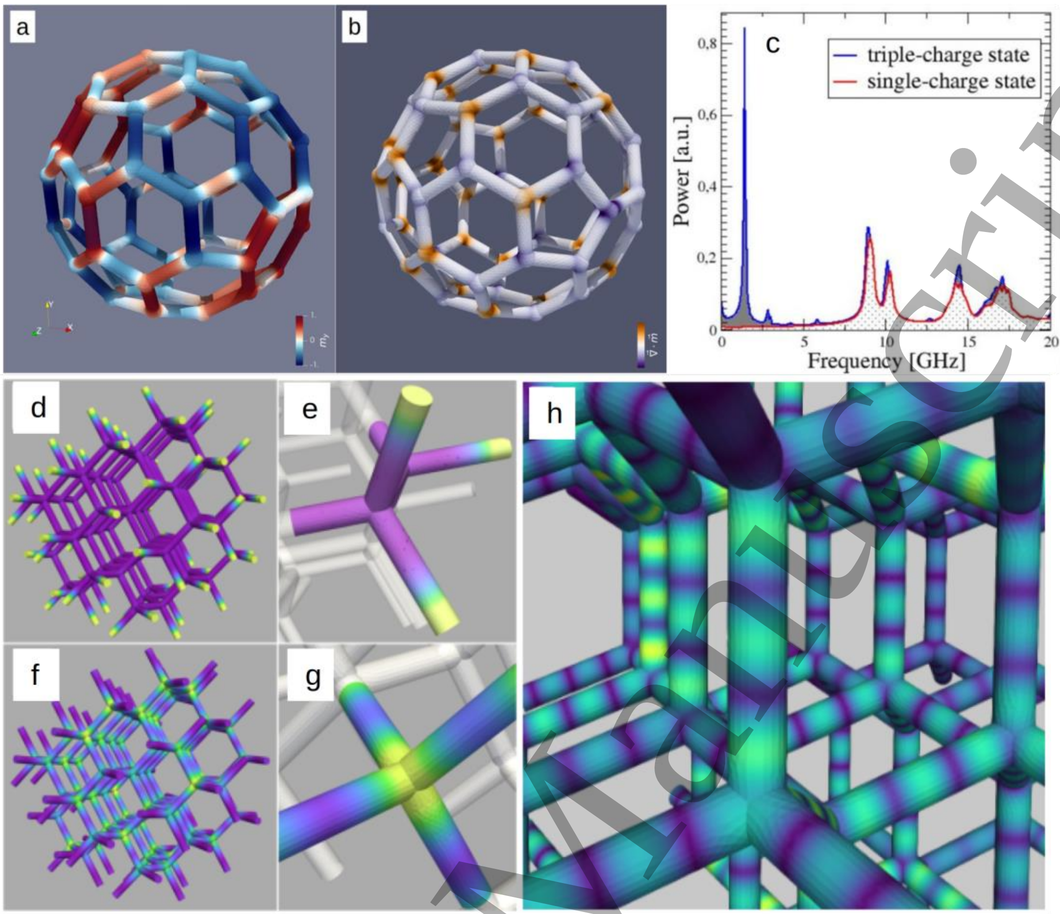

Figure 13.1. Simulated high-frequency magnetization dynamics in interconnected nanowire array geometries. The finite-element method allows for an accurate representation of the complex shape. Panels (a) and (b) show the static magnetization structure in a buckyball-type nanoarchitecture and its configuration-dependent oscillation spectrum, simulated in the time domain with the ringdown method (c) [Reproduced from [2], with the permission of AIP Publishing]. Panels (d)-(h) show various oscillation modes in a diamond-type artificial magnetic crystal developing at different frequencies. See [3] for details. Panels (d)-(h) are reproduced from [3]. CC BY 4.0.

To accurately model the complex shape of these nano-objects, simulations based on finite-element micromagnetics appear as the method of choice. Whereas finite-difference methods have been very efficient in simulating magnetic thin films, the finite-element method's capability to handle arbitrary shapes has become paramount in this emerging field of research. The situation is reminiscent of many other scientific domains, such as engineering, architecture, medicine, and aeronautics, where the need for accurate representations of the modeled geometry renders simulations with the finite-element method indispensable. In three-dimensional nanomagnetism, the commonly used micromagnetic simulation techniques based on the finite-difference method suffer not only from a lack of accuracy regarding the geometric representation but also from a loss of efficiency in the frequent case of samples with a low volume occupancy, such as interconnected nanowire arrays [2, 3, 4]. This is because the finite-difference method, contrary to the finite-element method, requires extending the calculation also to the non-magnetic part of the volume within the sample. A drawback of the finite-element method is that it entails larger numerical costs per degree of freedom, regarding both computation time and memory requirements, which results in a practical limit of the sample size that can be modeled. These aspects have been addressed, respectively, by employing GPU acceleration and hierarchical matrix methods, yielding a significant increase in speed and a drastic reduction of the memory requirements, thus allowing for large-scale micromagnetic finite-element simulations.

13.2 Current and Future Challenges

Three-dimensional nanomagnetic materials bear interesting potential for possible applications in magnonics. So far, phenomena related to spin waves in three-dimensional media, where networks of nanowires can act as wave guides and collective modes may develop, remain largely unexplored. A suitably chosen three-dimensional microstructure could significantly affect the generation and propagation of spin waves, resulting in magnonic metamaterials with tailored properties. In addition, the possibility to change the static magnetic configuration and thereby modify the material's magnetic high-frequency oscillations and spin-wave propagation could lend these objects unique reprogrammable features (see Fig. 13.1).

Simulation studies investigating such phenomena are traditionally carried out by modelling the oscillation of the magnetization, which is typically triggered by a field pulse excitation applied to the system. This "ringdown" method [5] consists in numerically integrating the Landau-Lifshitz-Gilbert (LLG) equation in time, where the effective fields acting on the magnetic moments need to be recalculated for each time step. Owing to the substantial number (hundred thousand or even millions) of discretization cells required, such time-domain micromagnetic simulations of the high-frequency dynamics of microscale magnetic 3D systems can be very time-consuming, even when accelerated by using fast numerical methods and exploiting the massive parallelism of graphical processing units (GPUs) [6]. In addition to the computation of the oscillatory magnetization dynamics over an extended period spanning several tens of nanoseconds – orders of magnitude longer than the individual sub-picosecond time steps required in the simulation, this traditional method also entails a complex Fourier domain analysis of the computed oscillations for extracting vital insights of the oscillations' frequency-dependence [5]. In the case of large-scale simulations, the significant computational resources currently required represent a practical limit in the possibility to explore the universe of possible dynamical response of 3D nanomagnets.

When simulating magnonic systems, an elegant alternative to the numerically costly time-integration of the LLG equation consists in exploiting two key features of the expected magnetization dynamics: the low-amplitude character of the oscillations around a stable equilibrium and their periodicity in time. Implemented in a mathematical framework known as the dynamical matrix method [7], these conditions make it possible to linearize the LLG equation and to solve it in the frequency domain, thereby obviating both, the need to perform the time-integration and to Fourier-analyze the results. Special frequency-domain simulation algorithms have been developed [7] which, by using a fully sparse, operator-based implementation, overcome the requirement to compute and retain a large, densely populated matrix within the computer's memory, which is the primary limitation of the dynamical matrix method, and thereby pave the way for large-scale simulations with low storage and computational costs [7].

13.3 Advances in Science and Technology to Meet Challenges

Micromagnetic frequency domain methods allow to investigate the oscillatory dynamics of 3D nanomagnets from two different perspectives. First, it is possible to characterize the small magnetization oscillations in the lossless (conservative, zero damping) limit around a given (spatially-inhomogeneous) micromagnetic equilibrium configuration as generalized eigenvalue problem for suitable operators related with the micromagnetic effective field. This formulation allows determining the normal modes (or, equivalently, the eigenmodes) of magnetization associated with their natural frequencies. One thereby obtains a discrete set of oscillation eigenmodes at specific eigenfrequencies, constituting a sort of "magnonic fingerprint" of the system. Second, by using the principles and techniques established for the eigensolver, one can construct a Matrix-Free Micromagnetic Linear Response Solver (MF-μLRS) for the calculation of the damped, steady-state oscillatory magnetization dynamics that occur when a weak sinusoidal magnetic field is applied externally. Perturbation methods combining the two above approaches allow for the efficient computation of any rf-driven magnetization dynamics for a given system, taking into account minor damping effects and arbitrary rf-field inputs [7]. Specifically, the ac steady-state magnetization response at the driving frequency, the power spectrum, and the absorbed rf-power can be analytically computed from the knowledge of a reduced set of magnetic eigenmodes.

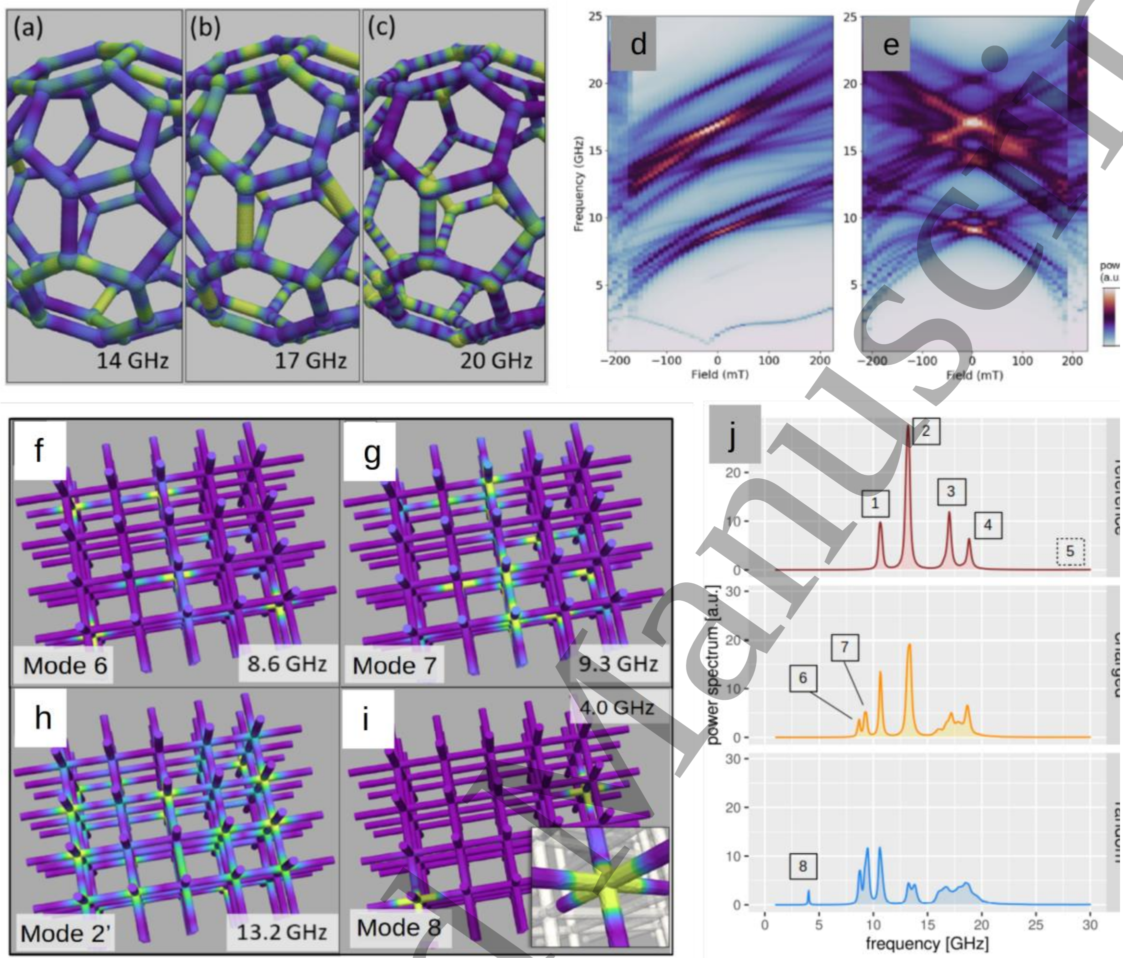

Figure 13.2. Advanced simulation methods in the frequency domain make it possible to conduct systematic simulations on the high-frequency magnetization dynamics, like spatial configuration and field dependence of resonance spectra for different magnetic oscillations in a buckyball geometry [see panels (a)-(d), reproduced from [8]. CC BY 4.0.], and to address the configuration dependence of the oscillatory dynamics in larger arrays of interconnected nanowires [see panels (f)-(j), reproduced from [4]. CC BY 4.0.].

These large-scale frequency-domain methods are being promisingly adopted for the detailed systematic study of the frequency-response of complex 3D nanomagnets, for instance as function of the external static field amplitude. As an example, one can see the magnetic-field dependence of the absorption spectrum for a buckyball structure (Fig. 13.2(d)-(e)) [8]. As expected, the field dependence of the absorption spectrum reveals highly complex pattern change in magnetization oscillations due to the reshaping of the (inhomogeneous) equilibrium ground state. Analogous investigations are performed on a nanowire array [4] and diamond-type artificial magnetic crystal [3] and some results are visible in Figs 13.2(f)-(j). The large-scale eigenmode approach can be also extended to investigate terahertz magnetization oscillations in the presence of inertial effects [9] and to build reduced-order models suitable to study nonlinear magnetization oscillations [10] eventually targeted to the realization of analog/neuromorphic computing.

13.4 Concluding Remarks

Within the broader context of three-dimensional nanomagnetism, three-dimensional magnonics is quickly emerging as a promising subdiscipline. Complex geometries, such as interconnected nanowire arrays, could act as programmable magnonic metamaterials with unique properties regarding their high-frequency response to external stimuli and spin-wave propagation. Reliable micromagnetic simulations are an essential prerequisite to investigating and interpreting magnetic oscillations and spin waves unfolding in such systems, where geometry details may significantly impact the magnetic response. Commonly used LLG-based finite-difference-based simulation techniques that have proven highly efficient in two-dimensional thin-film magnetism are poorly suited to address the challenges posed by the simulation of the oscillatory magnetization dynamics in these geometries. Recent developments combining GPU-accelerated finite-element techniques with frequency-domain formulation are the most promising numerical methods currently available to simulate such systems. These algorithms allow for a precise approximation of the geometry, owing to the finite-element discretization with unstructured meshes, and for a fast calculation of the oscillatory magnetization dynamics by circumventing tedious time-domain simulations and Fourier transforms. In combination with advanced sample fabrication techniques and suitable experimental imaging and analysis techniques, these numerical methods should prove particularly powerful for exploring the high-frequency magnetization properties of various largely unexplored three-dimensional nanomagnetic systems.

Acknowledgements

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14 – Advances and Future Directions of Three Dimensional Magnonic Crystals

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14.1 Status

Magnonics has the potential to make vast inroads into the on-chip data communication, processing and memory for spin-wave (SW) based computing. [1] Magnonic crystals (MCs) are the basic units of magnonic devices where the information carriers are SWs and a plethora of studies on two-dimensional and planar structures have been conducted in the last two decades. [2,3] Three-dimensional (3D) magnonic crystals have inspired the community due to their inherent analogy with the CMOS electronics where the third dimension aims to overcome the limitations of traditional two-dimensional integrated circuits by stacking multiple layers of electronic components.[4] This feature adds new functionalities beyond increase in storage capacity and also alleviates device connectivity which promotes neural networks, interconnectivity, controlled coupling between waveguides, unidirectional SW transport in non-reciprocal waveguides. In addition, it supports nonreciprocal edge mode and topological surface mode in magnonics. Recently, 3D nanomagnets have also *attracted much* interest because they exhibit unique magnetic properties due to their geometry, complexity of the spin textures, frustration and curvature induced effects. SWs can be scattered or reflected from spin textures (e.g, skyrmions, hopfions, domain walls etc.) while pinning centres can be used to trap and manipulate SWs.

SW spectra of 3D MCs were studied theoretically by plane wave methods in 2008, with ferromagnetic spheroids being distributed in cubic lattice (sc, fcc, bcc) embedded in a matrix of a different ferromagnetic material. [5] The magnonic gaps were found to occur at spontaneous magnetic contrasts that also depended on lattice type [Fig. 14.1 (a)-(d)] Experimental study of different types of 3D MCs have started recently. Kostylev *et al* presented first FMR measurements on 3D magnetic inverse opal structures [6] while large fcc lattices of magnetoferritin nanoparticles were investigated at low temperature using all-electrical spin-wave spectroscopy.[7] Time-resolved magneto-optical Kerr effect study of SW dynamics in 3D tetrapod structures fabricated by two-photon lithography (TPL) and electrodeposition showed the presence of a high-frequency standing SW mode and two lower frequency dipolar dominated mixed modes with nodal planes spreading along two mutually perpendicular directions [8]. Later in 2021, Sahoo *et al* demonstrated coherent SW modes in crescent shaped nanowires connected in 3D diamond bond lattice fabricated by TPL and thermal evaporation [9]. Two SW modes were observed in the experiment and simulation whose frequencies showed nearly monotonic variation with the applied field strength [Fig. 14.1 e,f]. They were found to be collective modes extending through the complex network while revealing spatial quantization with varying mode quantization numbers. More recently, a novel woodpile structure was fabricated using TPL defined 3D polymer scaffold conformally coated with a Ni shell using atomic layer deposition (ALD). This structure represents a 3D network of both horizontal and vertical interconnected and crossed nanowires [Fig. 14.1g]. BLS measurements of the SWs from the 1st and 2nd layer of the woodpile structure revealed about 10 GHz frequency discrepancy which was attributed to the surface and bulk SW modes of the system [Fig. 14.1 I,h]. Furthermore, angle and spatially dependent modes promise multi-frequency signal processing on 3D nanomagnetic networks [10].

A vast literature exists on the spin wave dynamics in two-dimensional (2D) magnonic crystals in multitude of systems like nanodot arrays, antidot arrays, nanorings, bicomponent magnonic crystals and quasiperiodic magnonic crystals [3,11]. Therefore, a direct parallel will be difficult to draw at this point as the literature on spin wave dynamics in 3D magnonic crystals are scarce. However, one may readily understand that the spin waves in planar structures are much simpler to measure and interpret as opposed to its 3D counterparts where the third dimension makes the spin waves more complex and difficult to measure. On the upside, the 3D systems allow the propagation and localization of spin waves in a variety of pathways, leading towards the development of more advanced and complex devices with the capability of storage and processing of larger data and performing more complex operations. For future applications it is desirable to have access to more control parameters for operation as well as ease of access. Long distance coherent propagation of spin waves is a challenge in 3D systems both from materials aspect as well complexity of structure. A trade off between materials, structures and external control parameters may lead to the desirable properties of spin waves as mentioned above.

14.2 Current and Future Challenges

The current and future challenges can be categorized into four groups: a. fabrication of high-quality samples, b. characterization of their SW properties, c. introduction of novel functionalities in the SW properties, and d. developing new simulation tools for SW properties as well as coupling mechanisms in 3D ferromagnetic architectures and devices.

The fabrication of 3D magnetic lattices faces a number of challenges. There are two main classes of

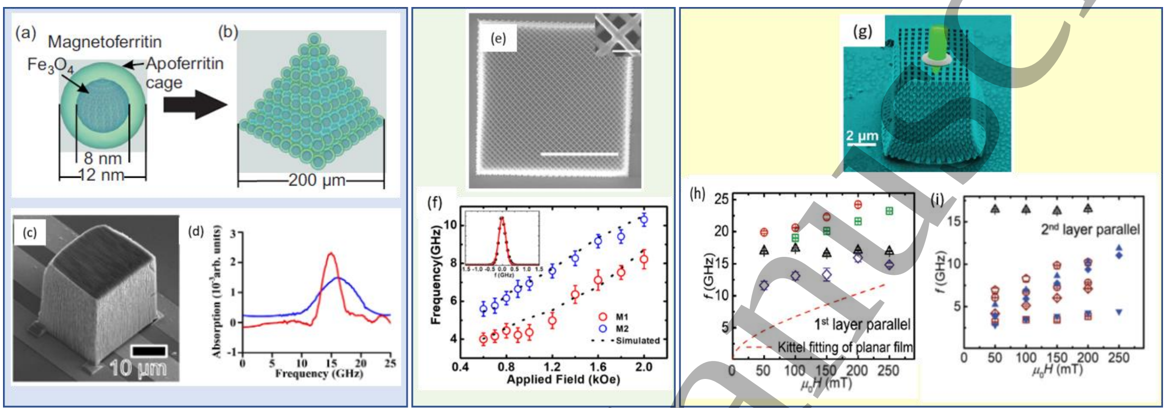

Figure 14.1. (a) Sketch of a single apoferritin cage with magnetite (Fe_3O_4) nanoparticle enclosed in its cavity. (b) Under appropriate conditions the nanoparticle arrange into a regular fcc lattice via self organization. (c) SEM image of a a cubic Fe_3O_4–ferritin crystal fabricated by FIB fixed to the two outer ground lines of a coplanar waveguide. (d) Microwave absorption spectra taken in an external magnetic field of 0.5 T applied in a direction perpendicular to the substrate (red line: 290 K, blue line: 5 K).[7] (e) SEM image of the full array of artificial spin-ice together and (f) the dependence of modes frequency on the applied magnetic field.[9] (g) SEM image of the 3D Ni nanowire nanonetwork (woodpile structure) and frequency dependence on modes when the applied field is parallel to the (h) first and (i) to the second layer of the woodpile structure.[10]

fabrication techniques: i. a direct writing technique of meta-organic precursor using focused electron beam induced deposition (FEBID) which is limited by the types of available materials (cobalt, iron and cobalt-iron alloys); and ii. physical or chemical coating of 3D nano-scaffolds fabricated by FEBID, TPL, self-assembly etc. However, line of sight deposition allows coating only to unit cell of lattice. Recent progress of conformal coating using electrodeposition and ALD has made possible to coat relatively large and complex structures like woodpile [10] or buckyball [12] structures. TPL allows additive manufacturing by depositing another functional material and the material assumes the TPL defined topography. One of the issues of coating nonmagnetic material by a magnetic material is that the resulting structure is magnetically hollow, thus making the modelling of such structures challenging [13]. Characterization of SWs in 3D MCs and 3D nanomagnetic structures have been primarily done by using BLS and time-resolved MOKE which are usually local techniques and therefore global information of coherent propagation and localization of SWs are often missing in such measurements. Besides, the optical technique can measure the SW dynamics only from the top surface of the 3D structure. Ferromagnetic resonance (FMR) is a global technique but is challenging to apply to 3D MCs due to difficulties in growing the 3D nanostructures on rf waveguides. Moreover, flip-chip technique is not trivial to use in this case without causing damage to the samples. The temperature dependence of the global SW dynamics is therefore difficult to study. The computation of 3D MCs and visualization of the complex 3D spin textures and SW mode profiles are also cumbersome. It is limited by computational resources as well as techniques to capture the SW dynamics and SW dispersion in complex structures. (Section 14) The agreement between experimental and simulated/calculated results poses great challenges. One aspect of modern magnonics is to introduce novel physical effects like spin-orbit coupling (SOC), voltage control, strain effects, topological effects, quantum coupling along with the SWs. However, introduction of these new functionalities requires introduction of additional materials and/or stimuli. For example, introduction of strain demands 3D scaffolds that can withstand strain. Addition of nonmagnetic layers for SOC requires techniques that support additive manufacturing.

14.3 Advances in Science and Technology to Meet Challenges

The study of 3D MCs challenges our fundamental understanding of the coupling, interference and propagation of SWs in multilevel waveguide networks and requires breakthroughs in the design and multilevel nanofabrication processes for the production of 3D MCs with enhanced precision and reproducibility over large dimensions. At the same time, the integration of 3D MCs into other technologies, such as spintronics and quantum computing, can open up new possibilities for information processing and storage, which could lead to the development of new types of sensors, signal processors, and other electronic components. In addition, miniaturized and more efficient energy transduction for the electromagnetic to SW signals need to be developed to reduce the power consumption and to create more sustainable and portable technologies. Finally, high-performance computers and sophisticated micromagnetic simulation techniques need to be developed to model and understand the complex SW dynamics of 3D MCs.

14.4 Concluding Remarks

Three-dimensional magnonics can unfold novel properties that are generally not observed in planar structures due to the inherent structural complexity and curvature induced effects.

It is therefore vital to explore three-dimensional magnonic crystals in order to achieve spin-wave manipulations and functionalities that are topographically impossible in two-dimensional systems. For example, the dependence of spin-wave properties and dispersion on the shape, size, lattice geometry of the magnonic crystal and on the strength and orientation of the external magnetic field will be an area of future interest as will the coupling mechanism between partially or fully coupled waveguides.

However, this will also face stern challenges due to the difficulties in fabrication of myriad of sample structures and their measurements as well as visualization using micromagnetic simulations. The introduction of novel functionalities like tailoring of magnetic charges and microstates in 3D artificial spin ice structure based upon 3D magnonic crystals can lead to exotic spin-wave properties due to the interplay between the spin waves and the magnetic charges/microstates (see also section 7). The utilization of other properties such as spin-orbit effects, quantum coupling, electrical control, strain etc. can also lead to advanced phenomena. Essentially, the discovery of novel effects specific to 3D structures will lead to new physics as well as the invention of devices such as high-density memory, logic and neuromorphic computing.

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15- 3D Magnonic networks and circuits

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15.1 Status

The development of magnonic devices has been very successful in recent years, providing a large choice of building blocks for increasingly complex magnonic networks and circuits [1]. These building blocks include, for example, magnonic waveguides, couplers, resonators, and entire logic gates like AND gates or magnonic half-adders. The first concepts and realizations of extended 2D magnonic circuits are currently being discussed and put into practice. In contrast, 3D magnonic circuits for data processing are still in a conceptional phase. One of the major goals pursued with their development is to resolve the connectivity bottleneck which would enable to develop highly interconnected spin wave circuits for brain-inspired computing.

3D magnonic circuits can be divided in two different classes depending on the flow of information. The simplest class, to which we refer as 2.5 D, is composed of 3D building blocks (e.g. waveguides) that are themselves arranged in a flat plane. These are, for example, systems of magnetic bilayers whose non-reciprocal magnonic dispersion relation can be used to form spin-wave insulators (see Fig 1b in section 12). Alternatively, 3D curved elements like magnetic tubes [2] (see Fig 1c in section 12) or curved topographical elements made of (or coated with) magnonic materials such as CoFeB alloys [3,4] (see Fig. 1a) or Yttrium Iron Garnet ($Y_3Fe_5O_{12}$, YIG) [5] can be utilized. All these 3D elements can show useful properties such as non-reciprocal magnon dispersion relations, which do not occur in flat single layer 2D systems. However, due to their usually small total thickness, the spin waves in these elements can still be approximately described using a 2D parametrization. This means that the wave vector ***k*** has only two non-vanishing components restricting the associated flow of information to two dimensions.

From a circuit perspective, the second class, the fully 3D magnonic systems where all three components of the wave vector are important for the information transport is very important (see Fig. 1b). It can be subdivided into systems consisting of multiple 2D layers (similar to the interconnect network in CMOS circuits) as depicted in Fig. 2, or volume systems where there is no fundamental distinction between individual dimensions. The latter approach could offer the greatest potential in conjunction with wave computing. However, it is also the least conceptually developed, as the flow of information in such a system is highly complex. In contrast to the layer-by-layer approach, such a system also requires an in-depth analysis of propagating spin waves in bulk materials. Here, the dispersion relations, propagation properties and nonlinear effects can significantly differ from the two-dimensional case of thin films. This is especially valid if the magnetostatic dipole-dipole interaction is relevant or if the bulk structure consists of a metamaterial, such as a 3D magnonic crystal.

Current experimental studies of fully 3D magnonic systems mainly investigate spin waves that are excited either thermally [6] or by a homogeneous GHz driving field [7]. The last approach would allow for an encoding of data in the time- or frequency domain, which could be relevant for simple data processing tasks like filtering, or specialized approaches like reservoir computing. However, to move towards extended logic circuits, excitation methods which allow for a spatially selective excitation of coherent, propagating spin waves are necessary. Thus, an important milestone to be accomplished is the experimental demonstration of coherent excitation and detection of propagating spin waves in a 3D structure.

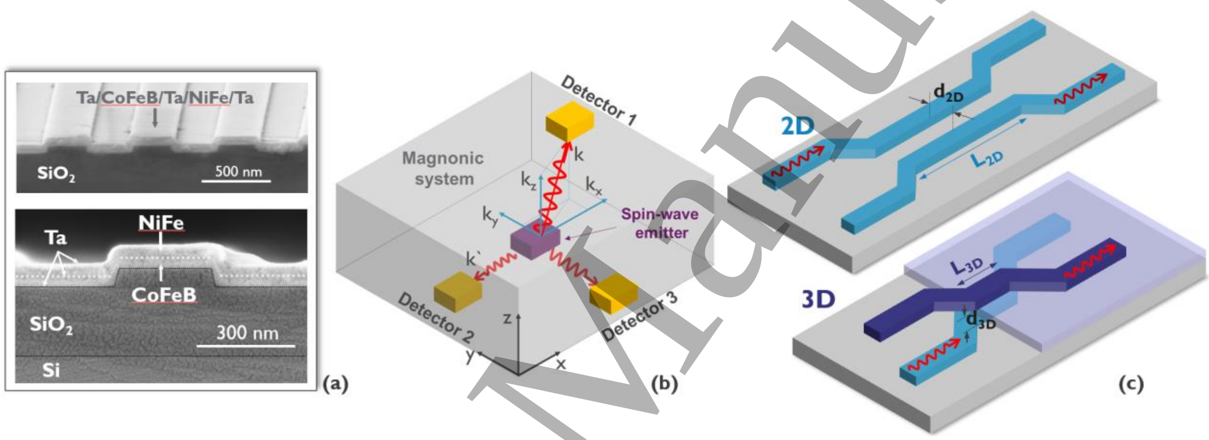

Fig. 1 (a) Scanning Electron Microscope images of a three-dimensional nanoscale CoFeB/Ta/NiFe meander structure. Reprint from Ref. [10]. (b) Schematic of a 3D magnonic system where the 3D character of the spin-wave wavevector selects the receiving detector. (c) Comparison of 2D and 3D magnonic directional couplers based on waveguides made from thin films. The most relevant parameter for miniaturization and energy consumption is the coupling length L. Due to the much smaller waveguide distance $d_{3D} \ll d_{2D}$ and the and the larger surface overlap in the 3D case, the coupling length L_{3D} is generally much smaller than L_{2D}. In 3D, the potential use of the exchange interaction to couple the energy between the two waveguides provides also a much more favorable scaling towards higher frequencies/smaller wavelengths.

15.2 Current and Future Challenges

A first challenge is to demonstrate the superiority of a 3D magnonic building block over its 2D counterparts. For example, magnonic directional couplers can be used for this purpose (compare Fig. 1(b)). Their 2D realizations already make it possible to cross spin-wave waveguides [8] for in-plane circuits. In contrast, 3D structures are required in electronics. The advantage of 3D magnonic couplers would be their increased coupling strength, as two thin waveguides can be placed precisely on top of each other with a very small distance between them. This achieves a short coupling length $L_{3D} \ll L_{2D}$ which is needed to transfer the wave between the waveguides, as recently confirmed in mesoscopic directional couplers with 200 µm waveguide width [8]. With more technically sophisticated coupling mechanisms such as exchange coupling, the coupling length can be reduced even further. From the estimation of the energy consumption of a device based on nonlinear directional couplers presented in Ref. [9], it can be concluded that for a given processing speed, the achievable energy consumption is proportional to the coupling length. According to this argument, the energy requirement of these systems could potentially be decreased by one to two orders of magnitude by transitioning to a 3D configuration, as the coupling lengths predicted by simulations are lower by about this factor. However, this only applies if the nonlinear coefficients in the 3D structures, for which no comprehensive calculations are yet available, are of the same order of magnitude as for 2D structures.

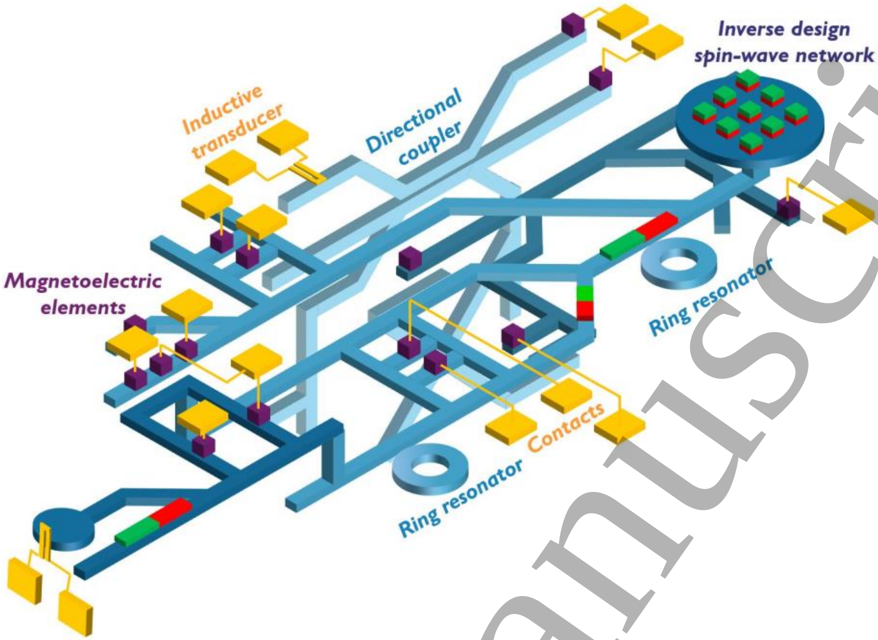

Fig. 2 Schematic view of a potential 3D magnonic network consisting of 2D layers connected by 3D directional couplers. Different elements like inductive transducers and magneto-electric elements (for spin wave excitation and phase shift), permanent magnets (for reconfiguration), ring resonators (filtering) and an area for inverse design are shown.

A further important milestone is to realize a magnonic system in which coherent propagation of spin waves is realized in all three dimensions. This requires embedding localized spin-wave sources whose dimensions are smaller than all three dimensions of the magnonic system. In a volume system, even without dissipation, there is a significant drop in the spin wave intensity I with the distance r $(I \sim 1/r^2)$. Therefore, the realization of guided magnon transport in 3D systems is particularly important. This can be realized either by special interference phenomena, such as spin wave caustics in 3D, or by three-dimensional waveguide arrangements as depicted in Fig. 2.

Numerous fundamental and technological advancements are still necessary to transform 3D magnonic circuits from concept to reality. In addition to the technological hurdles that must be addressed for device fabrication, analytical models for 3D systems and powerful simulation tools should also be developed. For example, the descriptions of 3D devices in terms of their linear and nonlinear spin-wave functionalities requires efficient micromagnetic solvers capable of simulating systems with hundreds of millions of cells. Furthermore, experimental characterization methods for the 3D propagation of spin waves [10] need to be further developed and accelerated. Currently, tracking spin-wave beams in 3D (bulk) architectures is extremely challenging.

15.3 Advances in Science and Technology to Meet Challenges

As previously mentioned, the (electrical) generation, manipulation and the (electrical) detection of spin waves propagating in 3D architectures is very demanding. The materials should be designed to ensure enough propagation length and desired magnetic properties, e.g., saturation magnetization, anisotropy, exchange constants, etc., for which spatial complex distributions could be expected. Advancements in deposition techniques now enable precise control over materials and stacks, with thicknesses reaching or even falling below 1 nm, as demonstrated for example in commercial Magnetic Random Access Memory devices. The anisotropy control was demonstrated by using different approaches, e.g., the deposition technique, the interface with different materials, ion implantation, etc. Nevertheless, the local control of the magnetic properties in 3D (bulk) systems requires further development. The patterning of spin-wave channels or waveguides and 3D architecture could benefit from the advances in CMOS technology where structures with feature sizes approaching 20 nm are close to being in production. Furthermore, waveguide patterning could also be achieved, by 3D laser writing into a semi-transparent magnetic material like YIG or by layer-by-layer growth like 3D printing techniques. Here, retaining low damping values of the single crystals materials and avoiding eddy current losses for the metallic materials is still a challenge.

The development of artificial intelligence and powerful computing systems will strongly enhance the design capabilities for complex 3D spin-wave architectures.

15.4 Concluding Remarks

Today, 3D magnonic networks and circuits represent primarily visionary concepts with the potential to achieve unprecedented connectivity in the GHz range. However, the challenges are substantial, covering both hardware, e.g., the realization of devices where coherent magnon propagates in 3D with low dissipation and low reflection, and circuitry concepts. The demonstration of 3D magnonic systems with advanced properties that go beyond those already shown in 2D remains to be established.

Acknowledgements

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16- Computation in 3D Nanomagnetic Systems

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16- Computation in 3D Nanomagnetic Systems

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16.1 Status

In 2012, about a decade ago, NAND Flash technology for solid-state mass storage devices became 3D. It came with relaxed constraints regarding bit area but simultaneously at the cost of increased fabrication complexity in transistor vertical stacking. Nevertheless, by 'simply' exploiting 3D stacking (hard-disc drives, compact discs, and Blu-rays remain stuck in 2D), a 100X improvement in 'areal density' has been achieved. Now, reaching the end of Moore's law of scaling, it is up to computing devices to follow this trend. While true 3D integration into mainstream MOS technology for general-purpose (digital) computing is not getting off the ground and, even worse, electrical losses in the complex wiring of modern ICs are increasingly proving to be a bottleneck, magnetic computing schemes with physically non-volatile state variables are promising. Computing in memory or memory as a computer, whatever you want. To give one example, perpendicular Nanomagnetic Logic (pNML), a 3D computing scheme for Boolean computation, offers intrinsically non-volatile computational states via stacked nanomagnets, as exemplified in Fig.1. Local engineering with ion beams 'tunes' the properties of the individual magnet, lateral dipolar magnetic fields provide next-neighbor computing capability, while vertical coupling fields enable signal transitions, and a global magnetic field clock provides switching power and synchronization [1-2]. A complete set of Boolean logic devices has been demonstrated, and validated system-level simulations suggest atto-joule dissipation per bit operation at CMOS-competitive data throughput. However, it's not the order-of-magnitude improvement we might be looking for, so as always, it's hard to beat CMOS where it's best: in high-performance digital computing. Still, for magnetic Boolean logic, there are some clear pathways following the approach of migrating similar to memory architectures from 2D to 3D, as mentioned above.

The other computing paradigm that is currently generating much attention amongst magnetism researchers is neuromorphic computing, a rapidly developing field that aims to draw inspiration from biological or neural network architectures to circumvent the increasing restrictions that thermodynamics, finite transistor size, and the von Neumann memory-processor bottleneck place on conventional CMOS digital Boolean logic.

Recent years have seen an explosion in the number and variety of magnetic neuromorphic computation schemes, from spin-torque nano oscillators to skyrmion systems and artificial spin ice nanoarrays. However, all existing schemes are two-dimensional, with no real forays of magnetic neuromorphic technology into 3D. This gives us somewhat of a blank canvas with which to write this roadmap, which is both freeing and challenging. The challenge being that we would like to identify routes forward which harness benefits that are uniquely available by moving magnetic systems into the third dimension and avoid the more generic advantages of 3D systems such as increased density, which are not specific to magnetic systems and appear in other 3D systems e.g. electrical & optical devices. Identifying such unique benefits of 3D nanomagnetic architectures before they have been demonstrated is a challenge, but an interesting one and we hope a somewhat valuable exercise.

16.2 Current and Future Challenges

A primary driver of neuromorphic computing is reaching the energy efficiency of biological brains. A human brain consumes around 20 W, while multi-GPU machine learning rigs regularly consume kW to MW. A huge part of this biological efficiency comes from the massively parallel interconnections of neuronal architectures. The human brain has hundreds to thousands of synapses per neuron depending on the region, allowing operations by a single neuron to efficiently inform many parallel subsequent operations at no additional energy cost. Existing computation is performed sequentially, with either repeated computations or repeated memory-processor shuttling operations generating huge additional energy costs.

Biologically-high degrees of interconnectivity are only remotely possible with the geometric freedoms of a three-dimensional system. We said we would try to resist the temptation of highlighting generic advantages of 3D, and increased parallel connectivity arguably touches on the generic appeal of increased areal density in 3D systems. Indeed, we are already seeing efforts to exploit this in 3D memristive crossbar arrays. However, nanofabricating conductive multilayer 3D interconnects between functional elements is still technologically challenging, and comes with additional Ohmic losses and thermodynamic challenges from increasingly dense wiring.

Something uniquely magnetic is exploiting the benefits of free-space dipolar coupling. Recently the effective strength of both static and dynamic components of dipolar coupling has shown capacity for substantial enhancement in 3D structures. While dipolar coupling has limited range, it is passive, natively 3D, and can couple both magnetostatic energy landscapes and GHz spin-waves/magnons. Functional designs such as the magnonic half-adder from the Chumak & Pirro group show the strong potential of such dipolar interconnects in 2D [3], migrating such designs to 3D offers unique benefits to magnetic computing that are not available in memristive or photonic architectures, the leading competing approaches. While the dipolar interaction may be thought of as somewhat weak relative to exchange coupling, recent work has shown that 3D architectures may be used to greatly enhance the effective intensity of dynamic dipolar coupling, with dipolar non-local ultrastrong magnon-magnon coupling demonstrated in a multilayered artificial spin ice [4] (fig 2a).

A key factor defining the versatility and computing power of neuromorphic systems is the range of available states. Recent work has shown that enriching the magnetic state space greatly enhances computing ability [5,6]. An attractive benefit of 3D magnetic systems is their expanded microstate spaces, including 3D-specific states. Examples include distinct bulk & surface magnon states [7], strongly-coupled hybrid chiral-collinear states [4], 3D solitons, and memristor-like multi-level states. Exploiting these states for enhanced neuromorphic computation is an enticing future direction. Gaining precise control over such expanded microstate spaces, both magnetostatically and in the magnon/spin-wave regime is a key future challenge for the field.

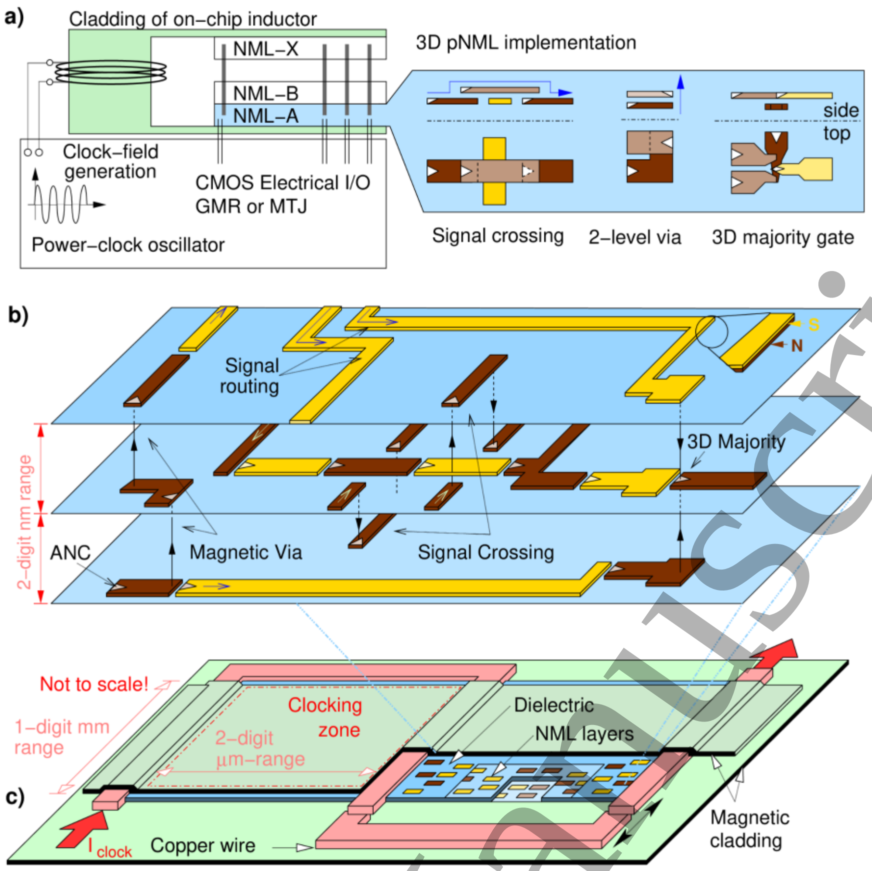

Figure 16.1. a) Nanomagnetic digital gates are stacked in the end-of-line technology of CMOS chips and powered/synchronized by a magnetic field. b) Ion-beam engineering to control domain reversal and domain-wall motion for signal routing is applied in a 3D stack. c) A highly permeable cladding combined with meander-like copper wires provides the magnetic field-clock. Adapted with permission of [1].

16.3 Advances in Science and Technology to Meet Challenges

In 3D nanomagnetic logic technology, care was taken not to connect each logic gate by electrical wires. The number of electrical I/O devices is orders of magnitude lower than the number of magnetic field-coupled gates. This comes at the cost that only a global magnetic field clock, integrated on-chip, could generate the needed bipolar field amplitudes for clocking in the two-digit MHz range. No efficient on-chip clock generator has yet been demonstrated, even if concepts can be adopted from on-chip inductors where soft-magnetic materials are applied for field concentration. However, it is an unsolved question: How can sandwiched computing elements be fed with electrical I/O structures and simultaneously clocked by a global magnetic field without significant interference (i.e., generated induction voltages)? Nanomagnetic logic relies on billions of simple binary 'states' laid out in vertically stacked planar geometry and coupled by local dipolar coupling fields for computation. Simple switches interconnected with inherent nonvolatility.

By contrast, neuromorphic systems draw their computing power from the complexity and rich state spaces of their functional elements. A distribution, stochastic or tailored, of functional elements may be engineered to produce usefully distinct responses from non-unique spatially distributed inputs, with greater potential density and diversity of these functional elements. Here, the challenge is to identify & engineer synergetic combinations of these functional elements such that computational power is maximised without overly-complicating fabrication, state-control and readout.

However, global magnetic field protocols (whether as clock or data-input) place limitations on both digital and neuromorphic schemes. Gaining enhanced field-free control of magnetic systems is a crucial future challenge for the field. Promising state-control solutions are being developed, including spin-wave [8] and all-optical [9] switching (fig 2b). These methodologies will require future refinement to tailor them 3D systems. While such field-free state control approaches are being studied in the lab, an appealing route to explore is all-optical magnetic writing using the spatial 3D laser control afforded by two-photon lithography systems such as those used in [7] and [10]. If these systems are already present in labs fabricating these 3D structures, enabling them to also provide proof-of-concept magnetic state reconfigurability is worth exploring (fig 2c).

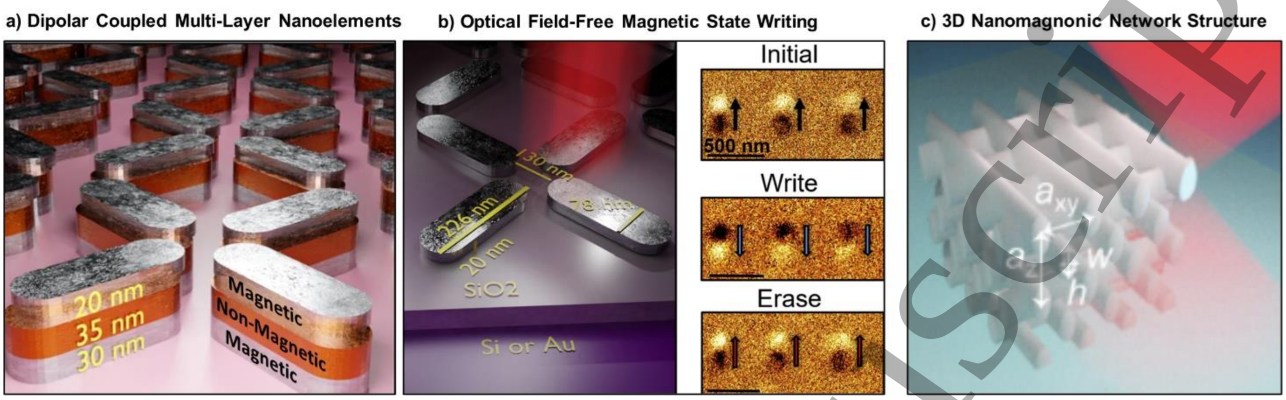

Figure 16. 2. 3D nanomagnetic systems and field-free writing approaching. a) Multilayered systems comprising strongly dipolar-coupled magnetic nanoelements may offer benefits for 3D magnetic computing, allowing information exchange and physical interaction between discrete nanoelements via the intrinsic dipolar field, with both static and GHz dynamic components. This may be a route to reducing or avoiding huge amounts of electrical interconnects in 3D nanostructures. Adapted from [4]. b) All-optical magnetic writing of nanostructures. 3D nanomagnetic systems require state control mechanisms that match the complexity of the 3D geometries if they are to be utilised as computing platforms. Mechanisms to write any nanomagnetic element, whether as a binary digital state or neuromorphic neuronal 'weight' are essential, particularly attractive if they avoid attaching electrical connections to every nanoelement. All-optical writing offers an appealing route forwards here, and has recently been demonstrated in 2D nanostructures. Adapted from [9]. c) Promising examples of 3D nanomagnetic architectures which should be explored for neuromorphic computing schemes include densely connected nanowire networks such as the example here from Guo, Grundler et al, adapted from [7]. Such systems offer both longer-range dipolar and strong short-range exchange coupling and the ability to host spin-waves/magnons which have been demonstrated as attractive potential information carriers in a host of 2D nanomagnetic neuromorphic computing schemes recently. The laser light here represents the two-photon lithography used to write the geometric structure, using such a voxel-focused laser with 3D spatial control could be an appealing future route towards 3D all-optical nanomagnetic writing similar to that shown in (b), albeit with technological challenges to be overcome.

Alongside state control, neuromorphic algorithms & training must be advanced to meet the challenges of complex 3D state spaces. Stochastic p-bit approaches are promising here, with designs harnessing non-local dipolar coupling of particular interest for 3D systems [11]. 'Digital-twin' and neuralODE models of 3D systems could play an increasingly important role here allowing a data-driven approach to modelling the complex 3D dynamics. However, it is prudent that such data-driven 'digital twin' models performed using neural networks are supported and confirmed by methods which have knowledge of the magnetic physics, such as micromagnetic simulation or analytical modelling.

Readout techniques must also be advanced to meet the challenges of dense 3D state spaces. Frequency-domain approaches are promising, capable of resolving some of the novel 3D states and behaviours [7] without requiring direct access to the hard-to-probe interior of 3D systems. We have seen 3D systems increase the frequency range across which modes are distributed [2,5], which is great for resolving distinct states and enhancing neuromorphic computation - but detection electronics will need to match these advances and develop low-power broadband detection.

16.4 Concluding Remarks

Monolithic fabrication of digital computing devices with 3D stacked nanomagnets is, in principle, similar to magnetic memory technologies and can naturally be integrated into the back-end-of-line of a CMOS process. Even though a complete set of devices has been demonstrated, there is a need for complex circuits, simultaneous integration of field-clocking structures, electrical transducers for I/O, and system-level benchmarking for real-world computing scenarios. Stochasticity of the magnetic states is likely to be one of the biggest hurdles since, in contrast to memory devices, every computing element must function reliably — memory is more forgiving.

Neuromorphic, fault-tolerant computing methods seem to offer routes around this stochasticity, particularly p-bit systems. Here, enhanced state-control, spatially distributed readout, and maturity of neuromorphic algorithms to match the enhanced complexity of 3D systems are likely to be the largest challenges in coming years.

Perhaps an interesting approach may be the development of 'hybrid' systems integrating deterministic Boolean magnetic logic with neuromorphic elements, implementing traditionally power-hungry digital-to-analog and analog-to-digital conversion via magnetic physics and offering crucial interfacing between neuromorphic processing and conventional CMOS systems via magnetic physics. It is unlikely that any computing system will be purely neuromorphic, and maintaining efficient compatibility with surrounding CMOS hardware is crucial to increase the prospects of industrial uptake of 3D magnetic computing technologies.

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17. 3-dimensional magnetic memory

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17. 3-dimensional magnetic memory

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17.1. Status

Figure 17.1 shows a conceptual diagram of the memory hierarchy in computers and other devices. It is well known that high-speed memories such as SRAM and DRAM are expensive and volatile memories with low recording density, while SSD and HDD are inexpensive non-volatile memories with high recording density. But there is a serious problem of triple-digit speed gap between DRAM and SSD. What is needed is a three-dimensional, high-capacity memory that is as fast as DRAM.

The so-called "race-track memory" proposed by IBM [1], which is based on the operating principle of current-driven magnetic domain wall motion [2], is expected to become a high-speed 3D large-capacity memory. The 3-D nature of the memory allows for a large amount of information per transistor, and the high-speed spin dynamics of the magnetic material makes it a high-speed memory.

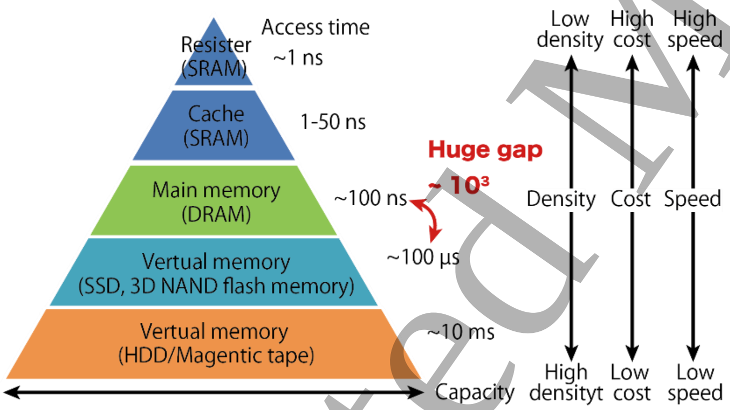

Figure 17.1. Conceptual diagram of memory configuration of computers.

Although the reduction of the threshold current density and the fast motion of the magnetic domain wall have been demonstrated, 3D race-track memory has not yet been demonstrated. The two problems that need to be solved to realize 3D race-track memory are how to precisely control the magnetic wall position and how to fabricate a 3D structure. As a three-dimensional structure to complete the magnetic wall position control, a 3D magnetic memory using artificial ferromagnetic materials as shown in Figure 17.2 has been proposed [3]. Each column has an alternating overlapping structure of a recording layer (green), which has large magnetic anisotropy and is responsible for bit recording, and a magnetic domain wall (DW) layer (yellow), which has no magnetic anisotropy and acts as an artificial DW layer. Simulations have confirmed that this 3D magnetic memory offers both high DW position controllability, thermal stability, and low power consumption [3,4].

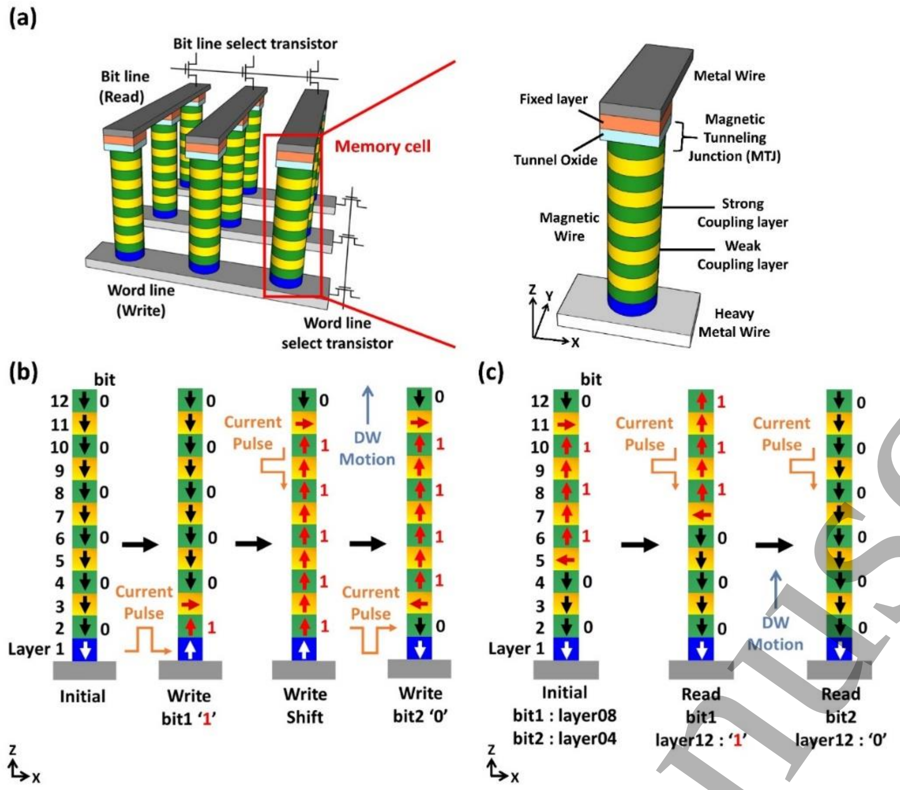

Figure 17.2. (a) Illustration of the vertical DW motion memory with artificial ferromagnets. Left panel: Memory array with vertically arranged word line and bit line. The columns within the array are the multi-bit memory cells constructed with artificial ferromagnets. Right panel: Detailed structure of a column. (b) Data writing scheme: A current is applied to the bottom electrode to invert the magnetization direction of layer 1 by spin-orbit torque. A DW is then generated in layer 3. The current is then passed through the column to shift this DW up to any DW layer. The next information is then written by inverting the magnetization direction of layer1 by applying a current to the bottom electrode. By repeating this operation, any bit sequence can be stored in the column. (c) Readout can be done in batches by reading out the information at the top MTJ while shifting all DWs upwards with a current [3].

17.2. Current and Future Challenges

A major challenge is how to fabricate a 3D structure like Fig. 2(a). Considering the aspect ratio problem of microfabrication even in the current 2D MRAM, it seems to be an extremely difficult task to fabricate a 3D structure like Fig. 2(a) with the currently available microfabrication technology. A possible solution is to first fabricate nanoholes to ensure a columnar structure, and then electrodeposit artificial ferromagnets inside the nanoholes. Using the latest technology developed for 3D NAND flash memory, it is possible to fabricate high aspect ratio nanohole arrays of tens of nanometers in diameter and several microns in length. The technology to create nanowires in such nanoholes by electrodeposition has already been established [5]. In fact, free-standing, interconnected metallic nanowire networks have been fabricated by electrodeposition into polycarbonate membranes that have been ion-tracked at multiple angles [6]. Such 3D interconnected magnetic nanowire networks can be expected to be integrated multistate memristors. To date, only field-assisted current-driven domain wall motion has been demonstrated in electrodeposited nanowires [7]. Therefore, current-driven domain wall motion in electrodeposited nanowires is another major challenge.

17.3. Advances in Science and Technology to Meet Challenges

In addition to the electrodeposition in nanoholes described above, several other techniques are being developed to fabricate 3D structures. For example, by using two-photon lithography followed by thin film deposition and direct writing by FEBID (focused electron beam induced deposition), 3D magnetic structures of almost arbitrary shape can be fabricated [8].

To realize 3D magnetic memory, in addition to the fabrication of 3D structures, technologies to visualize 3D structures at the nanoscale are required. In particular, a technology is needed to visualize not only the physical structure but also the magnetic structure at the nanoscale in three dimensions. The 3D magnetic tomography technique based on X-ray magnetic circular dichroism, combined with sample rotation control and a graphics processing unit to reconstruct spin structures inside bulk materials, is a promising technology of this kind [9].

In 3D magnetic memory, information is transported by domain wall motion, and Figure 17.2 assumes that the domain walls are driven by electric current. However, current is accompanied by Joule heating, which is a problem in terms of power consumption. Recently, a method to drive the magnetic wall by transferring the magnon angular momentum to the domain wall has been demonstrated [10]. The energy required to drive the domain wall is orders of magnitude less than that reported for metallic systems.

17.4 Concluding Remarks

With the commercialization of spin-transfer MRAM as an embedded memory, expectations for magnetic memory as a high-speed nonvolatile memory are rising. The development of high-speed, large-capacity 3D nonvolatile memory is essential for the future development of the information society. The key to the development of 3D magnetic memory will be the use of new materials such as artificial ferromagnets, artificial antiferromagnets, and artificial ferrimagnets, and the use of novel device fabrication techniques such as electrodeposition, two-photon lithography followed by thin film deposition, and direct writing by FEBID.

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18- 3D Spintronics

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18.1 Status

The expansion of nanomagnetism to three dimensions provides unique opportunities for the development of 3D spintronic technologies. Planar spintronic sensors and classical magnetic memories are made by thin film and standard lithography methods, restricting their functionality to the substrate plane. The usage of new spintronic devices, where the magnetic state is not confined to a single plane, will open a new era in spintronics, boosting storage and processing capacities, as well as device interconnectivity. It also has the potential to creating radically new computing architectures, driven by effects emerging with the change of dimensionality [1]. The paradigm shift of moving to 3D already initiated by CMOS [2], appears now on the hard disk drive industry roadmaps [3] and will strongly benefit emerging areas such as in-memory and neuromorphic computing (see section 16).

A natural way to go into 3D is via the usage of magnetic nanowires (see Figure 18.1(a-c)). The vertical racetrack memory is arguably the most iconic 3D spintronic device ever proposed (see section 17). Serving as an inspiration, many efforts are focused on exploring new effects associated to 3D motion of magnetic textures e.g. domain walls, skyrmions…, and recently magnons (section 15), via nanowire conduits. The most widespread approach followed so far to successful realize complex 3D nanowire circuits consists of the 3D printing of nonmagnetic scaffolds by FEBID or two-photon polymerization (see section 1 and section 2), followed by thin film deposition. These approaches are reaching a high maturity level [4] and can be extended to complex multilayered materials with functional interfaces. In particular, in-built gradients can induce strong automation of spin textures, a new mechanism proposed to unidirectionally interconnect spintronic devices located at different levels [5]. FEBID-written ferromagnetic 3D nano-bridges can connect planar circuits and present unconventional magnetotransport angular dependences, result of the complex magnetostatic vector fields present in a 3D geometry [6].

An alternative approach to direct 3D printing for the creation of 3D domain wall nanowire devices has been recently demonstrated, where a free-standing film is obtained by using an underlayer which dissolves in water, and can be positioned on top of pre-patterned substrates with humps of different radii. Using this methodology, synthetic antiferromagnet-based racetrack devices show a comparable performance to planar devices for moderate hump heights, demonstrating robust domain wall out-of-the-substrate motion at high speeds and low critical current densities [7].

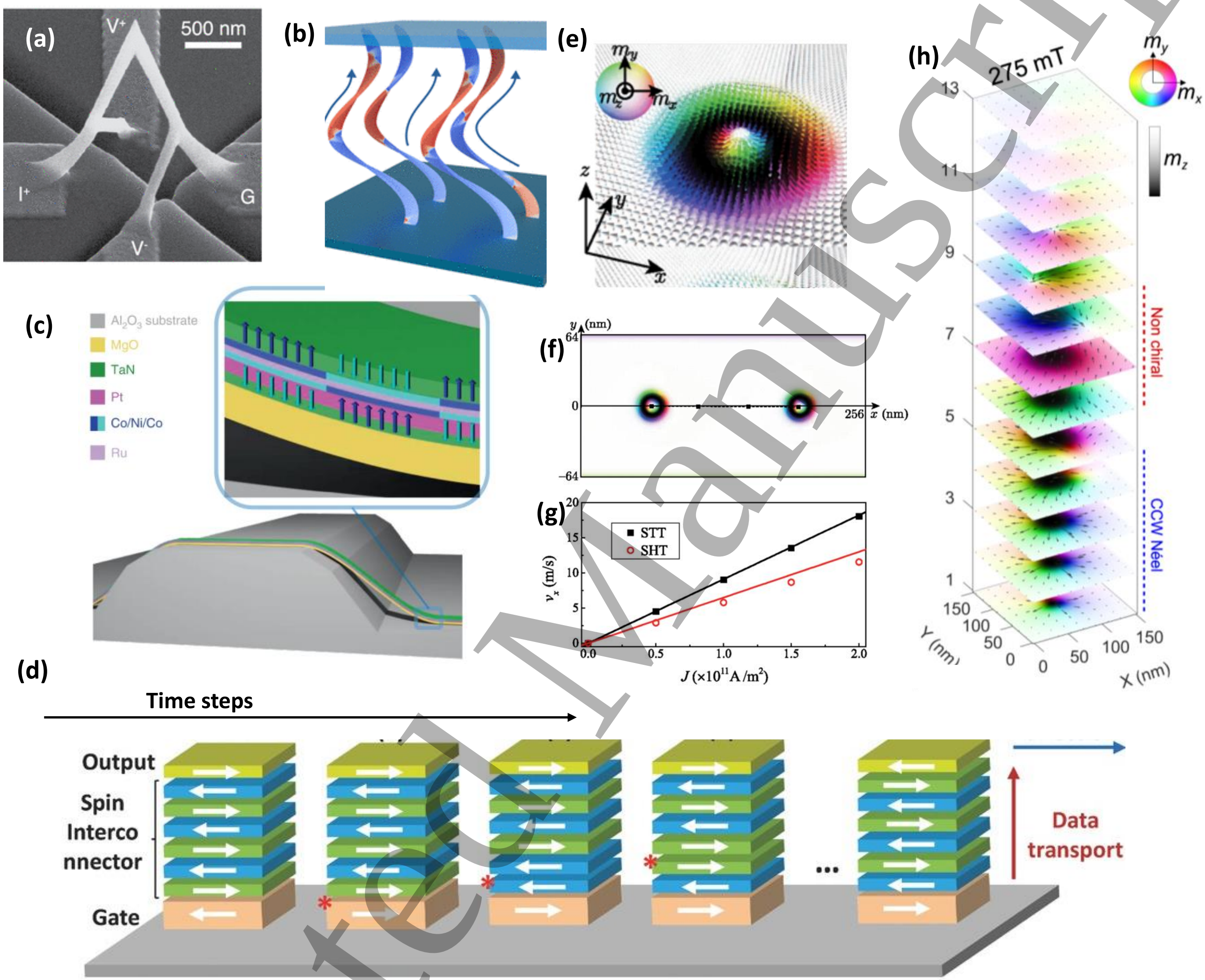

Figure 18.1. Recent works related to 3D spintronic devices. (a) Ferromagnetic interconnector directly printed by FEBID. Reproduced with permission from [6]. (b) Automotion of magnetic bits in between planes using interconnectors with in-built gradients. Reproduced with permission from [5]. (c) State-of-the-art racetrack memories formed by synthetic antiferromagnets, positioned on 3D pre-patterned substrates to achieve vertical domain wall motion. Reproduced with permission from [7] . (d) Concept for MRAM with vertical magnetic bit transfer functionality. Adapted with permission from [8]. (e-g) Theoretical study of current-induced motion of a Hopfion in a racetrack device. Adapted with permission from the American Physical Society [9]. (h) Micromagnetic simulations of a skyrmion cocoon formed along the height of ultra-thin magnetic multilayers, obtained by engineering the heterostructure energies. Reproduced with permission from [10]. Images (a, b, c, d, h) were originally published under CC-BY 4.0 license. https://creativecommons.org/licenses/by/4.0/deed.en

18.2 Current and Future Challenges

Direct-write lithography methods as described above can create circuits with arbitrary shapes, thanks to recently developed computational tools that enable 3D printing with nanoscale resolution (see section 1 and section 2) and [4]. However, these prototyping techniques, invaluable to investigate new physical phenomena, are not yet mature for high-throughput applications, and their integration with standardized lithography methods by the microelectronics industry will be very challenging. Multi-beam electron tools and patterning under cryogenic conditions, as well as advanced optical methods enabling multi-beam parallel patterning will improve speeds and could be a route to overcome related technical challenges. The introduction of cutting-edge spintronic materials (antiferromagnets, chiral ultra-thin multilayers…) into circuits with complex 3D geometries faces great challenges, but also incredible opportunities. Curved conduits with emerging magnetic energies are a first step in this direction (section 3 and section 11). Recent developments in chemical thin film deposition of ferromagnetic metals, and their integration in 3D scaffolds, is a promising future avenue. Chemical methods also provide fine control over the composition and microstructure of materials during growth, which combined with new methods for the patterning of 3D templates could create networks of highly interconnected nanowires. See section 1, section 14, and section 15 for further details.

Beyond spintronic devices based on new patterning methodologies, a seemingly more direct and robust path for 3D magnetic computing consists of employing multilayered heterostructures (Figure 18.1(d)), building upon the developments over decades of GMR and TMR based, sensing and memory devices (section 6). New schemes for 3D magnetic sensing based on spin-orbit torques in standard geometries have been already demonstrated[11]. The exploitation of shape anisotropy instead of PMA is one of the first realizations of pseudo-3D spintronic devices to achieve sub-20 nm MRAM devices[12]. A key aspect for real 3D functionality concerns the vertical motion of magnetic bits from one layer to the next, exploiting interlayer interactions [8]. Extending these ideas to current-driven schemes *e.g.,* for memory applications (section 17), overcoming negative effects in miniaturization due to strong magnetostatic interactions for large number of layers, and the continuum degradation of thin film properties for stacks based on hundreds of layers, are challenges ahead. Fine tuning of individual energies for each film on a magnetic stack can be also exploited for the formation of topological spin textures spanning across multiple layers, from coupled pairs of textures such as skyrmions to improve functionality, e.g., cancel the total skyrmion Hall effect, to the realization of topological 3D textures spanning across multiple layers such as hopfions and torons/cocoons textures (see Figure 18.1 (e-h) and a more in depth discussion in section 9). Moving such 3D spin textures efficiently under spin currents is still to be demonstrated, and their advantage for computing with respect to planar textures, will need to be thoroughly investigated. All these schemes will benefit from fundamental and technical recent advances, included the recent discovery of chiral interlayer spin interactions [13], and the development of magnetic characterisation and imaging methods; in particular, magnetic vector tomography at nanoscale resolutions (section 4, and section 5), key to resolve the static and dynamic vector dependence of these devices across its depth.

18.3 Advances in Science and Technology to Meet Challenges

3D spintronics is an emerging field with at the same time great opportunities and challenges. In a first stage, except for experimental works on cylindrical nanowires, the field was mostly theoretical and computational. For several years, a variety of new effects were predicted, from new types of domain walls to nonreciprocal motion of spin textures and magnons, emergence of chiral interactions due to nano-curvatures, etc. Few years ago, we entered a second phase, where experimental investigations became possible. The acceleration of this progress is very likely, due to the urgent demand for new forms of ultra-high storage and processing devices. We are now giving first steps in this second exciting phase, with theoretical predictions being finally tested, and new experimental discoveries taking place. Despite these advances, moving to higher Technology Readiness Levels (TRLs) remains challenging. Today the objective is to build novel demonstrators and confirm the expectation of such alternative functionalities based on 3D; tomorrow, the third phase will consist of conceiving and fabricating complex 3D spintronic circuits fully profiting of both the novel functionalities described above and the gain obtained by moving to the third dimension (see Figure 18.2). For this to happen, significant advances in robust, scalable manufacturing processes will be essential. Technical breakthroughs in techniques such as atomic layer deposition and large-scale parallel multiphoton lithography may become key drivers of this endeavour. Moreover, large clusters integrating multiple growth techniques—such as sputtering, atomic layer deposition, and pulsed laser deposition—along with in-situ characterization methods, will play a crucial role. When combined with robotics and AI for *e.g.,* automated diagnostics, these advances will enable the engineering of new, complex 3D spintronic devices.

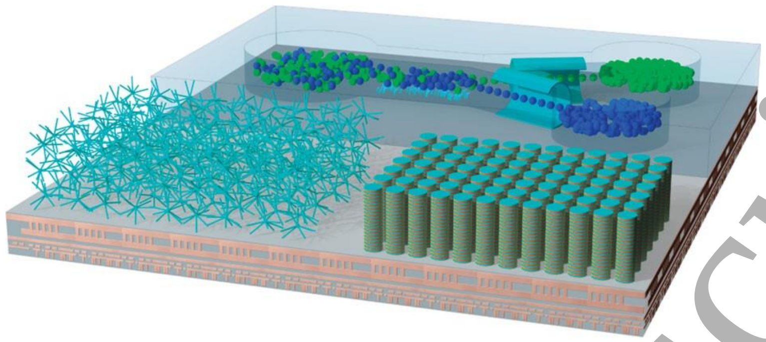

Figure 18.2. Futuristic view of a 3D spintronic multi-functional chip integrating different computing elements. This includes ultra-high data storage and memory devices, a microfluidic chip and a highly-interconnected network for efficient neuromorphic computations. Reproduced with permission from [1]. Original image published under CC-BY 4.0 license. https://creativecommons.org/licenses/by/4.0/deed.en

18.4 Concluding Remarks

3D spintronics, under the common umbrella of creating new spintronic devices with 3D functionality has several flavours, from 3D circuits with complex geometries, to magnetic multilayered based devices with magnetic functionality across the whole space, including 3D topological textures. Today Spintronics is bridging the gap with the microelectronic industry by conceiving new spin-chip integrating spins and CMOS [14]. Tomorrow, by giving access to the third dimension, complex circuits with completely novel functionalities and huge gain in performances will be achieved.

Acknowledgements

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