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Emerging materials in antiferromagnetic spintronics

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INTRODUCTION

The physics of compensated magnets (antiferromagnets, altermagnets, ferrimagnets at magnetic compensation, and synthetic antiferromagnets) is very rich, sometimes unique and unexpected compared to their ferromagnetic counterparts. New types of effects allowed in compensated magnets include ultrafast (THz) dynamics, pseudospin magnonics, (self-)compensated skyrmions, staggered topology, and compatibility with spin-polarized triplet superconductivity. The use of compensated magnets, therefore, constitutes a paradigm shift for the development of new spintronic components, beyond what is possible with the traditional ferromagnets. This special collection provides the reader with the latest material developments exploring the cutting-edge fundamental physics and promising applications of compensated magnets. It can be divided into seven thematic groups, each dealing with a current and fast-growing branch of the discipline.

TOPOLOGY

The compensated magnetic crystals featured in this special collection, with symmetry-imposed vanishing net magnetization, can be divided into the following three classes: (i) collinear antiferromagnets with opposite-spin sublattices connected by translation (t) or inversion (P) symmetry transformations. These symmetries generate electronic band structures with time-reversal (T) symmetry and spin-degeneracy across the whole Brillouin zone (at least in

the limit of vanishing relativistic spin-orbit coupling). (ii) Collinear altermagnets whose opposite-spin sublattices are not connected by t or P but are connected by rotation symmetry transformations. The corresponding (non-relativistic) electronic structures are characterized by a d-, g-, or i-wave spin-polarization that breaks T symmetry in the absence of a net magnetization. (iii) Non-collinear magnets with broken tT and PT symmetries and the compensation protected by symmetries combining crystal and spin rotation transformations. Unlike altermagnets, the corresponding electronic structures of these non-collinear magnets do not feature a spin polarization with a momentum-independent spin axis but rather a momentum-dependent spin texture. In analogy to altermagnets, the T -symmetry is broken in the (non-relativistic) spin-textured band structure of these non-collinear magnets despite the absence of a net magnetization.

The three classes of compensated magnets with their unique symmetries enrich a broad range of modern fields of condensed matter physics, including topological magnetism. The following Perspective¹ and three research articles^{2–4} give an overview and a detailed insight into prominent topological features of these three classes.

Reichlova *et al.*¹ review the topological signatures both in the real-space and in the momentum-space, discuss direct and indirect experimental measurement methods, and highlight the observed topological phenomena in prominent material examples of the collinear antiferromagnets and altermagnets, as well as of the non-collinear compensated magnets. Tschirner *et al.*² report a detailed

experimental study of the anomalous Hall effect in RuO_2 , which is one of the workhorse altermagnetic materials. Inbar *et al.*³ focus on their angle-resolved photoemission study on the topological features in a semimetallic collinear antiferromagnet GdSb , in particular on the control of the band inversion by strain. Finally, Wu *et al.*⁴ perform an experimental study of Mn_3Ge by combining anomalous Hall measurements and magneto-optical imaging of the nucleation and propagation of domain walls in this non-collinear compensated magnet.

SPIN TORQUES

For antiferromagnetic spintronic devices to be viable, one important criterion is the ability to manipulate the antiferromagnetic order in a reversible, controllable fashion. Electric control, in particular via current-induced spin torques, is the natural route to integrate antiferromagnetic media into practical devices. While some studies have pointed to the promise of spin-torque control of antiferromagnetic order, its true efficacy and the roles of competing contributions (such as thermal effects) remain to be fully elucidated.

The following three articles provide new insights into the control of magnetic order in antiferromagnetic films via current-induced torques. Shukla *et al.*⁵ numerically and analytically model spin-torque-driven dynamics of magnetic order in sixfold anisotropic Mn_3Sn . The authors investigate dynamics excited by a DC current that induces continuous oscillations of the magnetic order, as well as a pulsed current that switches the magnetization from one stable state to any of the other five stable states depending on the pulse amplitude and duration. This article also examines practical device considerations in detecting the magnetic order and accounting for Joule-heating effects. Xu *et al.*⁶ demonstrate that a current-induced torque can switch (11–20)-oriented epitaxial Mn_3Sn films with large thicknesses of up to 100 nm. This experimental study further shows that this switching behavior is unperturbed by external fields of up to 1.2 T and independent of the current axis orientation relative to the in-plane crystallographic direction. Amin *et al.*⁷ employ x-ray photoemission electron microscopy to show that electric current pulses can move 180° domain walls between distinct pinning sites in a patterned channel of CuMnAs . This controlled motion of 180° walls is distinct from prior demonstrations on 90° domain walls in CuMnAs .

Spin pumping is the reciprocal effect of spin torques and plays a key role in magnetization dynamics and spin-current generation. Spin pumping involving antiferromagnets is examined in two articles. Lim *et al.*⁸ experimentally study spin pumping in heterostructures incorporating thin films of Cr, an elemental antiferromagnetic metal. This work shows that Cr interfaced with Cu greatly reduces spin pumping and that this reduction does not originate from the antiferromagnetism of Cr. Tang and Cheng⁹ address the question of whether there exists a significant cross-sublattice contribution to spin pumping in antiferromagnets. This theoretical work indicates that such cross-sublattice spin pumping is absent in collinear antiferromagnets and very small in non-collinear antiferromagnets.

MAGNONICS

Just like ferromagnets, antiferromagnets can have dynamic excitations of their ordered magnetization structure, magnons,

which also can contribute to spin currents. However, the dynamics of magnons in antiferromagnets end up being fundamentally different compared to ferromagnets. The magnetization dynamics in antiferromagnets generally always includes a canting between the antiferromagnetic sublattices, which means that strong exchange interactions become important resulting often in a sizable energy gap (close to THz) for their excitation. In addition, unlike ferromagnets, magnons in antiferromagnets can have opposite chiral polarizations or even be linearly polarized. This results in new spin transport phenomena that cannot be observed in ferromagnets.

Two articles discuss antiferromagnets with canted non-collinear spin structures due to Dzyaloshinskii–Moriya interactions, and how magnons propagate diffusively after being incoherently pumped. Scheufele *et al.*¹⁰ observe in $\alpha\text{-Fe}_2\text{O}_3$ films that the magnon dynamics upon spin current injection via spin Hall effects from Pt contacts can be described by pseudospin rotation resulting in a magnon Hanle effect, and they investigated how this effect depends on thin film quality of $\alpha\text{-Fe}_2\text{O}_3$. Xu *et al.*¹¹ explore the magnon based spin transport in LuFeO_3 via thermal spin current generation with temperature gradients along different directions and conclude that the symmetries of their detected voltages from spin Hall effects in adjacent W are consistent with spin swapping, where the propagation direction and the polarizations of the spin currents are interchanged.

Finally, Biniskos *et al.*¹² use inelastic neutron scattering to determine how the magnon spectra change in Mn_5Si_3 upon magnetic field induced transitions of the spin structure.

IMAGING TECHNIQUES

The experimental observation of the magnetic configuration of compensated magnets, especially antiferromagnets and altermagnets, is challenging, since, overall, they lack a net magnetization. However, while meeting the challenge, the arsenal of techniques available for imaging compensated magnets has been considerably enriched and perfected. They include: (i) single spin magnetometry and relaxometry; (ii) magneto-optical techniques—such as x-ray optics for which magnetic circular or linear dichroism is combined with photoemission electron microscopy, Kerr microscopy, magneto-optical birefringence imaging, and second harmonic generation; and (iii) magneto-(thermo)electric microscopy—such as scanning tunneling and thermal gradient microscopy. A vast number of compensated magnets have been observed in those ways, irrespective of whether they are collinear or non-collinear in their ground states, are metallic or insulating (although some techniques are more suited for one type or the other), and host domain walls, vortices, or skyrmions. Three Perspectives in this collection deal with imaging of compensated magnets.

Zhou *et al.*¹³ provide an overview of techniques available for imaging compensated magnets and then focus on the optical method based on the magneto-optical birefringence effect and how promising this method is for real-time imaging of domains in antiferromagnetic materials. The magneto-optical birefringence effect is a quadratic effect arising from the difference in refractive indices when the light polarization is parallel or perpendicular

to the (sublattice) magnetization vector. As a result, the polarization direction of the reflected or transmitted light is rotated by an angle as it passes through an antiferromagnet with the Néel vector oriented in-plane, be it collinear or not. Despite its limited optical resolution (~ 300 nm), one of the main advantages of the technique is that compatibility with electric and magnetic fields enables real-time imaging of the dynamics of the antiferromagnetic configuration.

Kimura and Kimura¹⁴ review how nonreciprocal directional dichroism and related optical responses are used for the visualization of antiferromagnetic domains. The magneto-optical effect at play arises from the linear magneto-electric effect on electromagnetic waves, that is, a cross-coupling between electric and magnetic dipoles. As a result, in specific antiferromagnetic spin structures that break both space-inversion and time-reversal symmetries, the optical absorption can be changed by reversing the direction of light propagation or the sign of the magnetic order parameters, thereby enabling domain visualization. Despite its similarly limited optical resolution, temporal evolution of antiferromagnetic domains by use of pump-probe techniques is within reach.

Finco and Jacques¹⁵ review how single spin magnetometry and relaxometry techniques are applied to antiferromagnetic materials. Those are microscopy techniques that mostly use a nitrogen-vacancy center in diamond tips. In magnetometry mode, the combination of magnetic field sensitivity ($\sim \mu\text{T}$) and a high spatial resolution (~ 20 to 40 nm) makes it possible to return static snapshot images of the distribution of the magnetic field created by the uncompensated moments in an antiferromagnetic texture. In relaxometry mode, the coupling between the spin angular momentum of the nitrogen-vacancy center and the surrounding angular momentum—e.g., additional magnons when scanning across a texture—alters the spin relaxation time of the nitrogen-vacancy center, which can be measured.

OPTOSPINTRONICS

A critical advantage of antiferromagnets is their intrinsic ultrafast dynamics, but it is often not straightforward to trigger or study such dynamics with electrical means. For this reason, there is great interest in applying ultrafast optics to the studies and applications of antiferromagnets. With optical methods, it is possible to excite and probe ps-scale dynamics and induce novel spin-current phenomena.

The Perspective by Xiao *et al.*¹⁶ presents the spin photovoltaic effect in antiferromagnets. This effect allows for generating spin current in an ultrafast manner without any electrical contacts. The spin photovoltaic effect in antiferromagnets does not require spin-orbit coupling, unlike its counterpart in nonmagnetic materials. As such, it has potential implications for longer spin relaxation lifetimes in devices. Xiao *et al.* cover the basics of bulk and spin photovoltaic effects, particularly their symmetry constraints, and summarize recent studies of these effects in antiferromagnets.

Three research articles study ultrafast laser-induced responses in different types of antiferromagnets. Merte *et al.*¹⁷ present first-principles calculations of the photo-response properties of Mn_2Au , a widely explored antiferromagnetic metal. This study reports a large “photospin Hall effect,” the emergence of a transverse flow

of spins from a charge photocurrent. It also shows that symmetry breaking from a small canting of magnetic moments in Mn_2Au can produce a colossal chiral spin photocurrent. Besides the photovoltaic generation of the charge and spin currents, light can be used for the ultrafast manipulation of the magnetic interaction and dynamics in antiferromagnets. For example, Hortensius *et al.*¹⁸ investigate dynamic ferromagnetism in optically excited (photodoped) CaMnO_3 , a well-known strongly correlated antiferromagnetic manganite. They demonstrate that instead of a substantial chemical doping, a femtosecond laser pulse can induce an efficient photodoping to CaMnO_3 , resulting in the $\text{Mn}^{3+}/\text{Mn}^{4+}$ mixed valence states associated with the ferromagnetic double exchange interaction and, hence, a net magnetization. This experimental study reveals the inhomogeneous, patch-like nature of the ferromagnetic phase, rather than uniform spin canting throughout the material. In addition, Khusyainov *et al.*¹⁹ experimentally examine spin and lattice dynamics in CoPS_3 , a layered van der Waals antiferromagnet in which the Co^{2+} ion has a sizable orbital moment. They show that a femtosecond laser pulse can act as an ultrafast heater in CoPS_3 , resulting in the melting of the antiferromagnetic order and the effective changes of the orbital momentum of Co^{2+} , which triggers coherent THz phonons.

FERRIMAGNETS AND SYNTHETIC ANTIFERROMAGNETS

Ferrimagnets, consisting of antiferromagnetically coupled magnetic sublattices, made of different atoms or ions, and synthetic antiferromagnets, consisting of antiferromagnetically coupled ferromagnets, via the interlayer exchange coupling, offer tunable material platforms to explore some of the benefits related to magnetic compensation. In this collection, two Perspectives and one article address this topic.

In their Perspective, Caretta and Avci²⁰ review how domain walls speed up in insulating ferrimagnetic garnets. After an overview of domain wall dynamics, the authors present the advantages of compensated systems for domain wall dynamics, particularly with regard to spin angular momentum cancellation, which may differ from magnetic momentum compensation, and the role of Dzyaloshinskii-Moriya interaction and magnetic damping in achieving high domain wall speeds. How those parameters can be tuned in ferromagnetic garnets is extensively discussed.

Ko *et al.*²¹ evaluate the potential of ferrimagnets for hosting topological magnetic textures, in which chiral coupling plays a key role. In their article, the authors demonstrate chiral coupling between locally located orthogonal magnetizations in a single ferrimagnetic GdCo layer. Local out-of-plane magnetic moments arise from inhomogeneities and composition variations, while chiral coupling with the overall in-plane moments occurs via the bulk Dzyaloshinskii-Moriya interaction.

Wang *et al.*²² propose a Perspective in which they present why spin textures in synthetic antiferromagnets can be smaller, be faster, and propagate along a more rectilinear trajectory, due to compensation of the skyrmion Hall effect, than their ferromagnetic counterparts: all key advantages for their use in applications. An opening toward the construction of three-dimensional spin textures through the engineering of magnetic multilayers is also presented.

STRONGLY CORRELATED MATERIALS

Strongly correlated transition metal oxides host a broad range of phenomena of fundamental and practical relevance. In particular, the research on strong correlated antiferromagnetic oxides results in exotic new physics and promising applications. In this collection, two Perspectives were contributed in this topic.

Hu *et al.*²³ provide a comprehensive overview of the strongly correlated antiferromagnetic vanadates. They describe the physical properties of antiferromagnetic vanadates, including one-dimension (VO_2 and NaV_2O_5), two-dimension (VOCl), three-dimension (V_2O_3 , RVO_3 , AV_2O_4 , and VO), and single-stripe (Sr_2VO_4), which exhibit the strong correlations of spin, charge, lattice, and orbital degree of freedoms, giving rise to rich physical phenomena and potential spintronic applications.

In another Perspective, Yang *et al.*²⁴ review the antiferromagnetic iridates. The $5d$ electrons in Ir make the orbital momentum and spin-orbit coupling essential in iridates compared to that in $3d$ transition metal oxides, resulting in exotic antiferromagnetic orders. The authors introduce the recent progress on the reading of the antiferromagnetic orders in iridates via magnetoresistive responses and anomalous Hall effect and the writing using magnetic, electrical, and mechanical methods. They also propose several interesting directions, which may push forward the research on spintronic functionalities of antiferromagnetic iridates.

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Author Contributions

V. Baltz: Writing – review & editing (equal). **A. Hoffmann:** Writing – review & editing (equal). **S. Emori:** Writing – review & editing (equal). **D.-F. Shao:** Writing – review & editing (equal). **T. Jungwirth:** Writing – review & editing (equal).

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