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# Magneto–Optical Effects from Band Topology in Kagome Magnets

PhD Thesis Booklet

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# 1

# Introduction

The characterisation of different states of matter through the mathematical discipline of topology is a relatively young field of solid state physics. Its first success was the explanation of the integer quantum Hall effect in 1982 [1], while interest was significantly boosted after the year 2000 with the prospect of explaining the anomalous Hall effect in ferromagnets by both real– and momentum–space topological contributions [2, 3]. Subsequent theoretical efforts lead to the formulation of a periodic table of topological systems for gapped and gapless electronic phases depending on dimensionality and non–spatial symmetries [4], including topological insulators, Dirac and Weyl semimetals as well as topological superconductors. Although originally considered exotic phenomena, recent analysis estimates that more then 27% of known materials host some type of topological order [5]. Therefore, aside from potential technological applications in memory devices, spintronics and quantum computing [6, 7], investigating and engineering materials which realise these phases generates tremendous fundamental interest for understanding the interplay of single particle electronic properties, spin–orbit coupling, interacting (quasi)particles, magnetism and external stimuli such as strain and pressure or electromagnetic fields [8–10].

A material platform which provides an extremely versatile playground for the emergence and study of these phenomena is the kagome network. In this triangular lattice of corner sharing triangles, a plethora of exotic quantum states was theoretically predicted and experimentally observed, among them electronic and phononic flat–bands with enhanced correlations, charge density waves and topological superconductivity, as well as the quantum–spin–liquid state [8]. One of the earliest considered and most heavily investigated aspects is electronic topology under broken time–reversal symmetry, leading to a Chern insulator state and Weyl and nodal–line semimetals, for which the coupling with spin–orbit interaction produces highly unusual transport and optical properties that are the focus of this thesis.

Similar to the Haldane model for the quantum anomalous Hall state on the honeycomb lattice, a spin–chiral ferromagnetic kagome layer can also produce a quantised Hall conductance [11]. When such layers are stacked to form a 3D magnet, the topological properties can persist, resulting in a 3D topological–insulating or Weyl–semimetallic state [12]. This straightforward construction inspires the search for kagome lattice based materials realising these phases, although such realisations may be more complicated due to electronic correlations, finite–temperature effects or disorder. Central hallmarks of the presence of these states are the intrinsic anomalous Hall effect (AHE) and magneto–optical activity, which arise from the Berry curvature generated by the topological band structure in the presence of spin–orbit coupling. If generalised to finite frequencies, the two effects can be connected by the optical Hall conductivity which can be conveniently determined by measuring, e. g. the magneto–optical Kerr effect (MOKE) in magnetic metals. Since the latter is experimentally accessible by analysing the polarisation state of light reflected from the surface of a material, I use magneto–optical spectroscopy to investigate the topological properties of several kagome systems in this thesis.

# 2
# Objectives

The emergence of the AHE and MOKE in topological materials are closely related, as both require the breaking of time–reversal symmetry and finite Berry curvature in the band structure. But, while the AHE is a static transport quantity probing both the band structure and free carrier responses, the frequency dependence of MOKE and the derived optical Hall conductivity can give insights into the energy and (by comparison with *ab initio* calculations) also momentum distribution of topological bands in the Brillouin zone. In addition, the extrapolation of the broadband Hall effect spectra to the dc AHE can unambiguously separate the Berry curvature and scattering contributions to the off–diagonal transport, beyond the capabilities of magneto–transport experiments. Although an early pioneering study by Fang *et al.* [13] successfully applied this methodology to ferromagnetic $SrRuO_3$ and despite the effectiveness of the approach to identify the band origin of topological properties, the literature on magneto–optical spectroscopy applied to topological materials is rather slim.

Therefore, the goals of this thesis work are, on the example of the prominent class of kagome lattice based magnets, to (1) disentangle the AHE contributions of itinerant electrons and interband transitions to quantify the extrinsic and intrinsic proportions respectively, (2) from the multitude of bands for each material, identify the specific band structure features generating the intrinsic part to learn about the requirements for enhancing the anomalous response, (3) elucidate the role of spin–orbit coupling, (4) establish magneto–optical spectroscopy as a reliable tool to study topological band structures and to (5) monitor their variation through chemical pressure or external forces, e. g. doping or magnetic fields. The latter are predicted to provide an efficient control knob to vary the electronic band structure as both crystal symmetries and time–reversal may be altered by the applied field.

Along this line, I investigated the origin of the AHE in the archetypical kagome Weyl semimetal $Co_3Sn_2S_2$ and the room temperature kagome ferromagnet $Fe_3Sn_2$. In both materials, the large AHE may be associated with linearly dispersing bands which form nodal lines or are gapped except for Weyl points, while for $Fe_3Sn_2$, scaling relations also suggest a sizeable extrinsic contribution. Depending on the direction of the magnetisation, the number and position of these nodal features are predicted to change. Therefore, I analysed how high external magnetic fields manipulate the band structure in $Co_3Sn_2S_2$ with collinear ferromagnetic order, thereby demonstrating the applicability of magneto–optical spectroscopy also under such extreme external stimulus. In order to extend the approach to non–collinear magnetic structures, I finally investigated how the peculiar metamagnetic phases of the kagome spin–ice candidate HoAgGe influence its magneto–optical response to elucidate the coupling of electronic and magnetic degrees of freedom in this frustrated magnet.

# 3

# Methods

The experimental techniques necessary for magneto–optical spectroscopy I used in this thesis are broadband Fourier transform infrared (FTIR) spectroscopy for reflectivity and magneto–reflectance measurements and broadband magneto–optical Kerr effect spectrometers based on both grating and FTIR devices.

I performed the reflectivity experiments in Augsburg on a Bruker IFS66v and a Bruker Vertex 80v. The latter instrument was newly delivered during my PhD, so I participated in the initial setup to install the necessary cryostat and mirror optics to perform the broadband reflectivity measurements. With these two devices I measured reflectivity spectra over the range from $0.01 - 4\,\mathrm{eV}$.

The magneto–reflection experiments were performed during research visits at the LNCMI Grenoble in the lab of Milan Orlita, where a Vertex 80v FTIR spectrometer is coupled to either a resistive coil for measurements up to $34\,\mathrm{T}$ or a superconducting solenoid able to produce magnetic fields up to $16\,\mathrm{T}$. The sample holder could be configured for both Faraday and Voigt configurations and yielded magneto–reflectance spectra in a frequency range between $15 - 600\,\mathrm{meV}$.

The magneto–optical Kerr effect experiments were conducted at BME. Although the equipment was available before I started the PhD, both spectrometers had to be rebuilt completely for the experiments. I reset the optical paths and also refurbished the cryostat to operate properly at low temperatures. In order to extend the spectra to the far–infrared range, I developed a MOKE setup based on fixed polarisers coupled to a Varian 670–FTIR, which also required a service to allow stable operation. With these rebuilt and newly developed spectrometers, I measured the MOKE parameters from $0.025 - 4\,\mathrm{eV}$.

# 4

# Thesis Points and Publications

## 4.1 Thesis Points and Related Publications

Below, the major achievements of this thesis work are summarised in the form of the thesis points and the corresponding publications.

**Thesis Point 1**

On the itinerant kagome ferromagnet $Co_3Sn_2S_2$, I have measured the reflectivity spectra with in– and out–of–plane polarisation, and the magneto–optical Kerr effect (MOKE) spectra over a broad energy range from 25 meV to 3 eV. This required the development of a MOKE setup for the far–infrared spectral range, which I successfully realised by fixed polarisers for rotation measurements and a Kramers–Kronig constrained extrapolation of the ellipticity. Due to this extension, I could determine all elements of the conductivity tensor in this material for the specified energy range and capture the peaks caused by interband transitions of the gapped nodal line below 40 meV also in the off–diagonal component. Extrapolating to the dc values confirmed that the AHE in $Co_3Sn_2S_2$ has dominantly intrinsic nature caused by the nodal line and that Weyl points only give vanishing contributions. By calculating the frequency dependent Hall angle, I could show that these features are caused by an almost fully circularly polarised nodal line resonance, which additionally enhances the optical anisotropy favouring the out–of–plane conductivity.

F. Schilberth *et al.*, "Nodal line resonance generating the large anomalous Hall effect in $Co_3Sn_2S_2$", Physical Review B **107**, 214441 (2023)

**Thesis Point 2**

On $Co_3Sn_2S_2$, I have measured magneto–reflectance in Voigt configuration for two perpendicular field directions in the kagome plane, which result in the generation of a large number of Weyl points and a protected nodal loop. The resulting spectra show a spectral weight redistribution at low energies that follows the same trend as the in–plane magnetisation. I calculated the optical conductivity in field, which associates this redistribution to a peak caused by the nodal line resonance. Comparison to *ab initio* calculated spectra shows that the peak shift is associated with a narrowing of the SOC induced gap of the nodal line, while the large number of emergent Weyl nodes does not contribute significantly to the low–energy response.

F. Schilberth *et al.*, "Generation of Weyl points and a nodal line by magnetization reorientation in $Co_3Sn_2S_2$", arXiv.2408.03575 (2024) *submitted*

**Thesis Point 3**

In the kagome bilayer ferromagnet $Fe_3Sn_2$, I measured the broadband magneto–optical Kerr effect spectra between 50 meV–3 eV, which together with reflectivity data allowed me to calculate the conductivity tensor elements for the kagome plane. Again the far–infrared MOKE spectra enabled me to match the dc Hall effect to the optical spectra, revealing that the AHE in $Fe_3Sn_2$ is dominantly intrinsic above 100 K with a magnitude of $250\,\Omega^{-1}\mathrm{cm}^{-1}$. At lower temperatures, extrinsic scattering plays a major role. Due to the large number of bands close to the Fermi energy, the intrinsic AHE is produced by a large variety of transitions, but we could identify a distinct contribution from the helical nodal lines at energies below 0.2 eV.

F. Schilberth *et al.*, "Magneto–optical detection of topological contributions to the anomalous Hall effect in a kagome ferromagnet", Physical Review B, **106**, 144404 (2022)

**Thesis Point 4**

On the distorted kagome lattice compound HoAgGe, I have measured the anisotropic MOKE parameters as well as polarised reflectivity and magneto–reflectance for all crystallographic directions with magnetic field applied along the *b* axis. Due to the non–collinear magnetic order, we derived a formula for the Kerr parameters with anisotropic diagonal conductivity tensor elements, providing the basis for the analysis of high–field MOKE measurements. The broadband reflectivity measured between 10 meV to 2.2 eV shows strong anisotropy which translates to the optical conductivity along the stacking direction being larger than within the kagome planes. In the magneto–reflectance spectra obtained from 25 meV to 0.5 eV, the relative change of the reflectivity across the metamagnetic phases follows the magnetisation rather than the magneto–resistance for the in–plane response, suggesting a band reconstruction upon the magnetic transitions. The underlying changes of the electronic structure were analysed by evaluating the optical conductivity in magnetic field, which agree with preliminary *ab initio* calculations

F. Schilberth *et al.*, "Tracing Band Reconstructions across Metamagnetic Transitions in HoAgGe", *in preparation* (2024)

## 4.2 Full List of Publications

- F. Schilberth *et al.*, "Magneto–optical detection of topological contributions to the anomalous Hall effect in a kagome ferromagnet", Physical Review B, **106**, 144404 (2022)
- F. Schilberth *et al.*, "Nodal line resonance generating the giant anomalous Hall effect in $Co_3Sn_2S_2$", Physical Review B **107**, 214441 (2023)
- J. Ebad Allah *et al.*, "Optical anisotropy of the kagome magnet FeSn: Dominant role of excitations between kagome and Sn layers", Physical Review B, **109**, L201106 (2024)
- K. Vasin *et al.*, "Optical magnetoelectric effect in the polar honeycomb antiferromagnet $Fe_2Mo_3O_8$", Physical Review B **110**, 054401 (2024)

  F. Schilberth *et al.*, "Generation of Weyl points and a nodal line by magnetization reorientation in $Co_3Sn_2S_2$", arXiv.2408.03575 (2024) *submitted*
- F. Schilberth *et al.*, "Tracing Band Reconstructions across Metamagnetic Transitions in HoAgGe", *in preparation* (2024)
- V. Bader *et al.*, "Rotational disorder and its impact on the spin–1/2 triangular antiferromagnet $Na_2BaCo(PO_4)_2$", *in preparation* (2024)

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