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Review on the PhD thesis of Felix Schilberth Title: Magneto-Optical Effects from Band Topology in Kagome Magnets

The concepts of topology have been revolutionizing condensed matter physics. In particular, magnetic topological materials, such as magnetic topological insulators, magnetic Weyl semimetal and skyrmion materials, are of great current interest; therein the non-trivial band topology coupled with the intrinsic magnetism arising from electron correlation brings about unprecedent magneto-transport properties and related functions.

The motivation of this thesis by Felix Schilberth is to experimentally elucidate the nontrivial electronic band topology in some of prototypical kagome-lattice magnetic seimimetals characterized by gapped Weyl nodes. For this purpose he exploited the broadband magneto-optical spectroscopy, including Magneto-reflectance (MR) and magnetooptical Kerr effect (MOKE). The itinerant magnets with a Kagome lattice are expected to be a gold mine for intriguing quantum phenomena arising from frustrated magnetism and topological spin textures.

The theme of this thesis turns out to be right due to the proper choice of both methodology and target materials; Felix Schilberth showed in this thesis that the magnetooptical spectroscopy can play a decisive role in concluding that the large anomalous transport properties observed in these compounds are due to non-trivial band topology, in particular the gapped Weyl nodes near the Fermi level.

Thesis Structure

This thesis is composed of seven chapters and appendix. The first chapter of this thesis is excellently written, summarizing the basic concepts of electronic topology and their implications for the kagome magnets. Chapter 2 is devoted to the methodology of magneto-optical spectroscopy. In particular, the infrared and far-infrared (down to 25meV) MOKE spectroscopy on the bulk single crystals is essential for the spectroscopic resolution of the near Fermi-level band topology responsible for the emergence of the large anomalous Hall effect in DC transport. The description is concise and concentrated, while including the state-of-the-art MOKE methodology, appearing as an excellent review paper chapter for the broader readership. The remaining chapters in this thesis

present the experimental results and analyses/discussions for selective kagome magnets; Chapter 3 (MOKE) and 4 (MR) for the archetypical kagome Weyl semimetal Co₃Sn₂S₂, Chapter 5 (MOKE) for the room temperature kagome magnet Fe3Sn2, and Chapter 6 (MR) for the distorted-kagome spin-ice material HoAgGe. Chapter 7 is devoted to the summary of this thesis, in which the experimental achievements and conclusions are clearly summarized as Thesis Point 1-4.

Thesis Content

The main achievements by Felix Schilberth in this study and the evaluation of those by this Reviewer are as follows:

- (Chapter 3) He conducted broadband magneto-optical (MOKE) spectroscopy on the nodal line semimetal Co₃Sn₂S₂. He successfully determined all non-zero elements of the optical conductivity tensor, including both longitudinal(σ_{xx} and σ_{zz}) and transverse (σ_{xy}) components, in the photon energy range from 25 meV to 3 eV. At around 40 meV, he clearly identified a transition with giant optical Hall angle $\Theta_H(\tan\Theta_H = Re(\sigma_{xy}/\sigma_{xx}))$ close to the fundamental limit, which additionally enhances the optical anisotropy $\sigma_{zz} / \sigma_{xx}$. By comparing with first-principles calculations, he assigned this resonance to transitions between the lower and upper bands of the spin-orbit-coupling (SOC) gapped nodal line. While one preceding infrared (down to 80meV) MOKE study of Co₃Sn₂S₂ was reported, his advanced MOKE study down to 25meV is essential to finally catch the clear and quantitative feature of the gapped Weyl nodes, showing the interband resonance around 30-40meV. This presents the first definite experimental evidence to show that the anomalous Hall effect in Co₃Sn₂S₂ has the dominantly intrinsic nature caused by the gapped (~30meV) Weyl nodal line.
- (Chapter 4) On the basis of the one-to-one spectral correspondence between σ_{xx} and σ_{xy} established in Chapter 3, he exploited the MR to investigate the change in the electronic band topology near the Fermi energy when the magnetization of Co₃Sn₂S₂ is slanted from the magnetic easy axis (perpendicular to the Kagome plane) by applying a high magnetic field up to 34T. He attributed a field-induced low-energy shift of the giant magneto-optical resonance (~30meV) to a narrowing of the SOC induced gap of the notal line. This interpretation is consistent with the results of first-principles calculations. Such a magnetic control of band topology as observed in the SOC induced gap excitation on the notal line is an outstanding achievement in the field of topological materials.

- (Chapter 5) Next, he investigated the MOKE response for a room-temperature kagome magnet Fe₃Sn₂, which is predicted to have the helical nodal line. He found the smooth connection between the low-energy optical Hall effect and the DC AHE in Fe₃Sn₂ above 100K as well as in the case of Co₃Sn₂S₂. On the other hand and also importantly, he found a large discrepancy between the low-energy optical Hall effect and the DC AHE at lower temperatures in Fe3Sn2. This points to the extrinsic (*e.g.* skew scattering) contribution to the AHE dominant at low temperatures where the DC conductivity (carrier scattering time) increases. As far as this Reviewer knows, this is the first spectroscopic evidence for the energy-scale difference between the intrinsic and extrinsic mechanisms of AHE.
- (Chapter 6) He investigated the MR spectra of itinerant distorted-kagome lattice magnet, HoAgGe, as a function of the magnetic field, across a sequence of metamagnetic transitions through the magnetization plateau states. He observed various low-energy (<0.5eV) interband transitions and demonstrated large magneto-reflectance sensitive to the band-structural changes with the magnetization.

Conclusion

In my view, the quantity and quality of this work is on an outstanding level of a doctoral thesis. The candidate's advanced magneto-optical spectroscopy technique, as exemplified by far-infrared MOKE and high-field far-infrared MR, is extremely high, enabling him to experimentally approach to the gapped nodal-line close to the Fermi level under magnetic fields which is believed to be the microscopic origin for the intrinsic mechanism for the large anomalous Hall effect. His careful and insightful experiments, analyses and discussions on some representative kagome-lattice magnets, as well as his deep understanding of underlying physics of the topological magnets, leads to the present high-level scientific achievements. His scientific productivity is evidenced by his publication record, as listed in Chapter 7. In relation to the thesis, he has published two Physical Review B papers and submitted one manuscript, all with him as the first author.

Based on the above appraisals, I accept the results described in this thesis as the original achievements of Felix Schilberth and strongly recommend this outstanding research for public defense.

Questions

- (1) The far-infrared MOKE down to 25meV is to be appreciated. As the candidate described, however, the sensitivity of the FIR rotation is smaller than for PEM-based technique in the mid-IR region. Nevertheless, the present far-infrared MOKE appears as an excellent tool to identify the gap excitation on the gapped Weyl nodes in Co₃Sn₂S₂. Can the author show the substantial example how well the raw FIR region MOKE data can be connected to the PEM-based data in the energy region of 60-100meV?
- (2) As for Figs.3 and 4, the ~30meV resonance in the optical longitudinal and transverse conductivity is a most important signature for the gapped Weyl node in Co3Sn2S2, while it is less pronounced in the calculation. Why is this difference? What is the role of electron correlation (proposed in the thesis) to pronounce the spectral intensity of the peak?
- (3) In relation to the discussion of Re σ_{xy}(ω → 0) in comparison with the dc AHE data, the author ascribed the discrepancy to the coexistence of intrinsic AHE (appearing in Re σ_{xy}(ω → 0)) and extrinsic AHE (appearing only in the dc data). I think that this interpretation is plausible and that the FIR magneto-optical estimate of the intrinsic (Berry-curvature origin) AHE is meaningful. The latter extrinsic effect, e.g. skew scattering, is quite sensitive to the scattering rate of the charge carrier, generally becoming important when σ_{xx} exceeds 10⁶S/cm, therefore its disappearance with increase of temperature is also reasonable. Can the author be more quantitative about the estimate of this important distinction between intrinsic and extrinsic AHE effect for Fe₃Sn₂ crystal used here? For example, what is the temperature dependence of DC σ_{xx} and DC σ_{xy}? What is the energy scale of the extrinsic AHE, or would it be absorbed in the Drude response?

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