Response to the questions of Dr. Levente Rozsa

on the dissertation

Magneto–Optical Effects from Band Topology in Kagome Magnets

by Felix Schilberth

Thank you for your thorough and very positive evaluation of my thesis. Below, I answer the questions you put forward point by point.

(1) In a lattice model, the scalar spin chirality is defined by $\chi_{123} = \mathbf{S}_1 \cdot (\mathbf{S}_2 \times \mathbf{S}_3)$, which is nonzero if the three local magnetic moments $\mathbf{S}_i$ span a solid angle. The projection of this chirality to the plane of the lattice sites determines the Berry curvature which corresponds to a fictitious magnetic field $\mathbf{b} = \sum_{i,j,k} \mathbf{S}_i \cdot (\mathbf{S}_j \times \mathbf{S}_k)\, \hat{\mathbf{n}}_{ijk}$, where $\hat{\mathbf{n}}_{ijk}$ is a normal vector to the triangle spanned by the three lattice sites i, j, k [1, 2]. In a continuum model, this emergent magnetic field takes the form

$$b_p = \frac{\hbar}{2} \epsilon_{pqr} \hat{\mathbf{m}} \cdot \left(\frac{\partial \hat{\mathbf{m}}}{\partial q} \times \frac{\partial \hat{\mathbf{m}}}{\partial r} \right) \tag{0.1}$$

where $\hat{\mathbf{m}} = \mathbf{m}/|\mathbf{m}|$ is the direction of the magnetisation vector and ϵ_{pqr} the total antisymmetric tensor with the spatial coordinates p, q, r [3, 4]. It is possible to derive topological invariants from this quantity, as e. g. the skyrmion number can be computed by integrating $\mathbf{b}$ over the crystal plane.

(2) There are two aspects which are relevant to address this question. First, if the electronic structure of the compound is well represented by the *ab initio* calculations, the band structure and optical optical spectra do not require extra tuning parameters. This was the case for e. g. $Co_3Sn_2S_2$ where a standard density functional theory approach was sufficient to satisfactorily reproduce the experimental data with a single particle picture. On the other hand, for Fe_3Sn_2, the Hubbard U had to be included to achieve a reasonable agreement. Although the value used agrees with other experiments, e. g. angle resolved photoemission spectroscopy, some tuning was necessary. For HoAgGe, the effects of the $4f$ electrons need to be considered which also goes beyond a straightforward *ab initio* approach but may be overcome by a sophisticated tight binding model. So different theoretical approaches could in principle reproduce the same diagonal conductivity spectrum if tuning is possible.

The second aspect is specific to the magneto–optical methods used. Spectral weight in the diagonal conductivity $\sigma_{xx(yy,zz)}$ can arise from a combination of various transitions which makes the assignment of spectral features difficult. E. g., the step feature in Fig. 5.5(a) of the thesis, which in our decomposition is a cumulative response from many interband transitions, was assigned to the response of a Dirac dispersion based on an oscillator model in a different publication [5]. This ambiguity is a central reason why in the present work, the main claim on the topological properties of bands is based on the analysis of the optical Hall conductivity σ_{xy}. Following Eq. 1.32, spectral weight to this quantity can only arise if the bands carry Berry curvature. So a remarkable resonance in this spectrum as observed in $Co_3Sn_2S_2$ requires the presence of a topological band structure feature at the corresponding energy and with significant joint density of states. Therefore, although the literature regarding the analysis of such spectra in the context

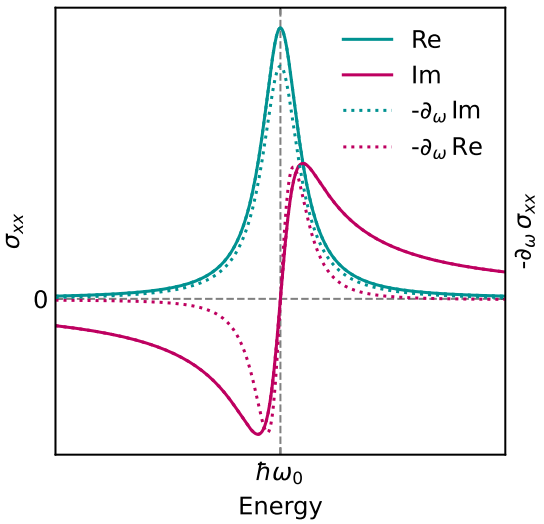

Figure 1
Real and imaginary part of a Lorentzian oscillator in the optical conductivity σ_{xx}. For comparison, the negative derivatives of the respective quantities are also shown to highlight the similarity of the lineshapes.

of topological materials is not extensive, I believe that the simultaneous agreement of the calculated spectra with both diagonal and off–diagonal conductivity makes a strong case towards the topological nature of the band structure.

(3) The ratio and its preference of the in– or out–of–plane anisotropy can be deduced from Ref. 6. In Table 1, the authors list the spin–orbit matrix elements between the d orbitals as defined by $\langle m, \sigma | \mathbf{L} \cdot \mathbf{S} | m', \sigma' \rangle$. In this reference, $m = 1 - 5$ is used as an index for the orbitals $d_{yz}, d_{xz}, d_{xy}, d_{x^2-y^2}, d_{3z^2-r^2}$ and σ corresponds to the spin. These elements are determined by applying the angular momentum and spin operators on the d orbitals in the basis of spherical harmonics, which introduces the polar and azimuthal angles η and ϕ listed in the table. To determine the in– or out–of–plane anisotropy, η should be set to 90° or 0°, respectively. For $m = m'$, these matrix elements are always zero. The expectation value obtained by taking the absolute square will result in the ratios stated in the paragraph of the thesis.

So indeed, it is not straightforward to deduce which combination of orbitals favours which type of anisotropy. Additionally, since in correlated metals, the band structure at the Fermi level has hybridized orbitals, this generally gives a qualitative description. However, in $Co_3Sn_2S_2$, the nodal line is dominated by the in–plane $d_{xy}, d_{x^2-y^2}$ orbitals, as is shown in the projected band structure in the supplementary Fig. E5 of Ref. 7.

(4) Eq. 1.33 shows the special case how to determine the dc anomalous Hall conductivity from the Kramers–Kronig (KK) relation of the spectrum of Im σ_{xy}. The general KK integrals for the off–diagonal conductivity read

$$\text{Re } \sigma_{xy}(\omega) = \frac{2}{\pi} \mathcal{P} \int_0^\infty \mathrm{d}\omega' \frac{\omega' \,\text{Im } \sigma_{xy}(\omega')}{\omega'^2 - \omega^2} \tag{0.2}$$

$$\text{Im } \sigma_{xy}(\omega) = -\frac{2}{\pi} \mathcal{P} \int_0^\infty \mathrm{d}\omega' \frac{\omega \,\text{Re } \sigma_{xy}(\omega')}{\omega'^2 - \omega^2}. \tag{0.3}$$

Of course, the derivative cannot be used as a substitute for the proper KK transform, but the statement in question mentions a peculiarity of the shape of the real and imaginary parts. To illustrate this, I show the optical conductivity σ_{xx} of a Lorentzian oscillator, e. g. an infrared active phonon, in Fig. 1. Here, the real part is the dissipative response, hence it shows a peak

at the resonance frequency ω_0. In the imaginary part, determined through the KK relation, the lineshape shows an S–structure at the resonance. Up to a sign change, the lineshapes of the two KK related spectra therefore resemble the respective derivative in frequency also shown for comparison. This rule of thumb also works for other types of resonances and excitations as may be observed in Fig. 1.21 of the thesis. For the spectra in Fig. 5.3 (or other figures where two KK related quantities are plotted together e. g. 3.2, 3.3 and 5.2), this means that whenever a peak occurs in one of the spectra, the other should show a slope originating from the S–shape, but an explicit zero crossing at the peak position is not required by the KK relations. Since the real and imaginary parts of the KK quantities may be determined independently, e. g. the rotation and ellipticity during the magneto–optical Kerr effect measurements, this observation can serve as a quick check whether the measurements yield a reasonable result.

(5) In the ground state of HoAgGe, the magnetic order is non–collinear, but still coplanar according to Ref. 8, so the solid angle spanned by any three spins is zero. Applying a magnetic field along the b axis keeps the spins in the plane due to the strong Ising nature of the rare earth moment. So for the present set of data, real–space topology does not seem to play a role. However, in the Supplementary Material of the above reference, the magnetisation for field along the c axis is shown which implies a gradual canting of the moments which opens the possibility for scalar spin chirality. Unfortunately, no Hall measurements were reported for this configuration, so I cannot tell whether this creates a topologically non–trivial response. For the coplanar magnetic structures however, Ref. 9 reports significant anomalous Hall effect with unusual hysteresis between the plateau phases, suggesting non–trivial electronic band topology in this compound.

Nevertheless, if real–space topology contributes to the static Hall response, it is not unreasonable to expect signatures also in the optical spectra, as e. g. additional contributions to the magneto–optical Kerr effect in the mid–infrared range were found in the skyrmion lattice phase of Gd_2PdSi_3 [10]. These results indicate that the generation of topological Hall effect from real–space Berry curvature is not necessarily limited to the action of the emergent magnetic field on conduction electrons, as it may also modify the band structure and influence the magneto–optical response.

(6) In the context of chapter 6, the term “quasi–symmetry” refers to an operation that transforms the lattice of HoAgGe onto itself, but is not a space–group symmetry. This unusual concept was introduced in Ref. 9 for a combined operation of a two–fold rotation and a “distortion reversal” of the kagome lattice. While for the model introduced in this reference, that operation also leaves the band structure invariant, it does not preserve the transport response. Hence Neumann’s Principle is violated by this transformation, which would not be the case for a space–group operation.

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