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Thermal Hall Effect of Topological Triplons in $\text{SrCu}_2(\text{BO}_3)_2$

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The quantum magnet $\text{SrCu}_2(\text{BO}_3)_2$ is a physical realisation of the Shastry-Sutherland model - a two-dimensional square lattice, solved to have a ground state of singlets on diagonal bonds. Recent theoretical work suggests that the material should play host to a topological phase of triplons, which may manifest a thermal Hall effect. However, within the measured experimental resolution, we report that there is no thermal Hall signal of the predicted magnitude. This is possibly due to triplon-triplon interactions playing a more significant role than anticipated in the temperature range under investigation.

KEYWORDS: Thermal Hall, Triplon, $\text{SrCu}_2(\text{BO}_3)_2$

1. Introduction

The discovery of the quantum Hall effect stimulated a revolution, bringing topology to the forefront of condensed matter physics [1]. More recently, the field was reinvigorated by the prediction, and subsequent experimental realisation, of the class of materials termed topological insulators [2, 3]. In such materials, it is the electronic band structure which is topologically non-trivial. Recently however, there has been a drive to investigate topological phases of bosons.

Bosonic systems offer both practical and conceptual advantages over their fermionic counterparts. For one, bosonic quasiparticles are weakly interacting, as they are electrically neutral, and may therefore be better suited in realising dissipationless edge states. Conversely, in the absence of Fermi statistics, the weak interactions must be the origin of any topological phenomena. Thus, a mastery of simpler bosonic systems may pave the way to a better understanding of topological effects in correlated fermionic systems [4].

Topological phases of phonons [5], photons [6] and magnons [7] have already been theoretically predicted, with the latter two having also been experimentally realised [8, 9]. This investigation concerns a novel proposal - that the quantum magnet $\text{SrCu}_2(\text{BO}_3)_2$ should exhibit a thermal Hall effect, carried by a topological phase of triplons [10, 11].

2. Theoretical Understanding

$\text{SrCu}_2(\text{BO}_3)_2$ is a remarkable material in that it is a physical manifestation of an exactly solvable model, known as the Shastry-Sutherland lattice [12]. This is a two-dimensional square lattice of spin-1/2 moments, which, under certain conditions, has been solved to have a ground state of singlets on diagonal bonds. As such, the material has garnered decades of research [13], with experimental findings such as the spin-gapped ground state [14], localised magnetic excitations [15] or magnetisation plateaus [16], all adequately explained by theory [17].

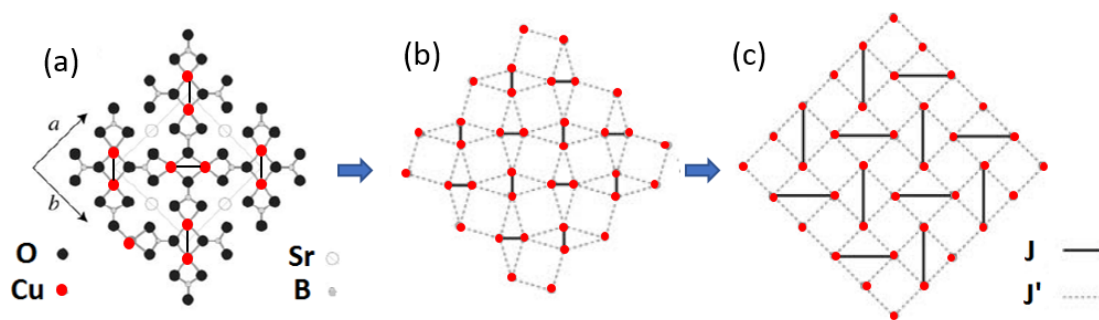


Fig. 1. (a) the atomic structure of each two-dimensional layer in $\text{SrCu}_2(\text{BO}_3)_2$ (where the Sr and CuBO_3 planes are displaced), (b) the dimerised magnetic structure of the same layer, with the spin-1/2 moments on the Cu sites having been isolated, (c) the theoretical Shastry-Sutherland lattice. Figure amended from [13].

With reference to Figure 1, if $J'/J < 0.68$ then the singlet ground state is energetically favourable [18]. In $\text{SrCu}_2(\text{BO}_3)_2$, this ratio has been experimentally determined to be 0.635 [19], satisfying the constraint and implying that the lowest energy magnetic excitation should be the promotion of a single dimer into the triplet state. Early inelastic neutron studies found these triplet excitations to be extremely localised [15], as predicted within the Shastry-Sutherland model. However, a slight corrugation of the layers below a structural transition at 395 K [20] allows for Dzyaloshinskii-Moriya interactions on all bonds. As a result, the (aptly named) triplons acquire a small dispersion and, importantly, are imbued with topological character.

In order to quantify this topological character, McClarty et al [10] constructed a model to include all symmetry-allowed couplings between nearest and next-nearest neighbour spins, building upon the elegant, but simplified, work of Romhanyi et al [11]. This allowed for an extremely accurate (in comparison to inelastic neutron data) determination of the triplon band structure, from which the Berry curvature of each band could be calculated. Finally, the Berry curvature can be integrated over the two-dimensional Brillouin zone to give the Chern number of each triplon band - an integer topological invariant.

In quantum Hall systems, the transverse electrical conductivity is quantised corresponding to the Chern number of successively filled orbits. However, the analogue for bosonic systems - the transverse thermal conductivity - is instead dependent on the magnitude of the Berry curvature in thermally occupied parts of the band, and is therefore not quantised [21, 22]. This can be seen in the calculated thermal Hall signal for $\text{SrCu}_2(\text{BO}_3)_2$ [10, 11] which forms the basis of this investigation, and is shown in Figure 4 alongside experimental data.

3. Experimental Procedure

Single crystals of $\text{SrCu}_2(\text{BO}_3)_2$ were grown with 99%-enriched boron-11 by the optical floating zone technique at 0.25 mmh^{-1} under 3 bar oxygen pressure. The measured sample was approximately $2.5 \times 2 \times 0.25 \text{ mm}$ in size. Shown in Figure 2 is the experimental setup. Thermal links to the sample have been made using DuPont 6838 silver epoxy. The heat bath comprised a 1 mm silver wire, soldered onto the OFHC copper sample stage. Heat links to the three CX-1050 thermometers, as well as to the strain gauge heater, were made with $25 \mu\text{m}$ gold wire.

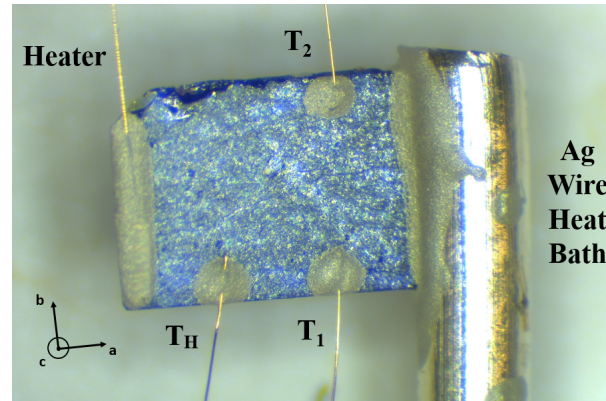


Fig. 2. Microscope image of the sample, with thermal contacts positioned such that the longitudinal and transverse thermal conductivities could be measured simultaneously. The axes shown refer to the crystallographic axes.

Heat was applied parallel to the a -axis and a magnetic field was applied parallel to the c -axis. As shown in Figure 2, the setup allowed simultaneous measurement of the longitudinal and transverse thermal gradients - $\Delta T_x = T_H - T_1$ and $\Delta T_y = T_1 - T_2$. The thermal conductivities could then be calculated from

$$\frac{1}{wt} \begin{pmatrix} Q \\ 0 \end{pmatrix} = \begin{pmatrix} \kappa_{xx} & \kappa_{xy} \\ -\kappa_{xy} & \kappa_{xx} \end{pmatrix} \begin{pmatrix} \Delta T_x/L \\ \Delta T_y/w \end{pmatrix}, \quad (1)$$

where t is the sample thickness, and L and w are the distances between the contacts T_H-T_1 and T_1-T_2 respectively. In order to correct for any potential misalignment of the contacts T_1 and T_2 , the data has been antisymmetrised ($\Delta T_y^{AS} = [\Delta T_y(+B) - \Delta T_y(-B)]/2$, where B is the applied field). To ensure accuracy of calibration, data was taken using temperature sweeps at a constant field. The same setup was also successfully used to detect a thermal Hall signal of a similar magnitude in a different material, as described in the Appendix.

For every data set, both the longitudinal and transverse thermal conductivities were calculated. The former provided a number of useful checks. Typical longitudinal conductivity data for $\text{SrCu}_2(\text{BO}_3)_2$ is shown in Figure 3. This exhibits a double peak structure as a function of temperature, as well as a reduction in magnitude of the lower temperature peak with increasing field. Both of these features have been observed in previous studies [23], and both may be explained by a resonant scattering of phonons from triplet excitations. The thermal conductivity therefore provided a basic check that the material was behaving as expected. In addition, in order to improve precision, the sample heater was set to provide the maximum amount of power which would not lead to significant heat leakage - a constant thermal conductivity ensured that this was the case.

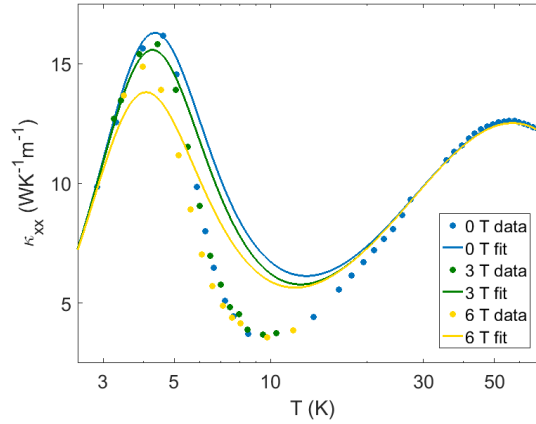


Fig. 3. Thermal conductivity data for $\text{SrCu}_2(\text{BO}_3)_2$. Points represent experimental data and lines are a fit to the model described in [23].

4. Experimental Results and Discussion

Shown in Figure 4 is the transverse thermal conductivity data for $\text{SrCu}_2(\text{BO}_3)_2$, with the theoretically calculated curves shown for reference. Within the experimental resolution, there is no appreciable thermal Hall signal.

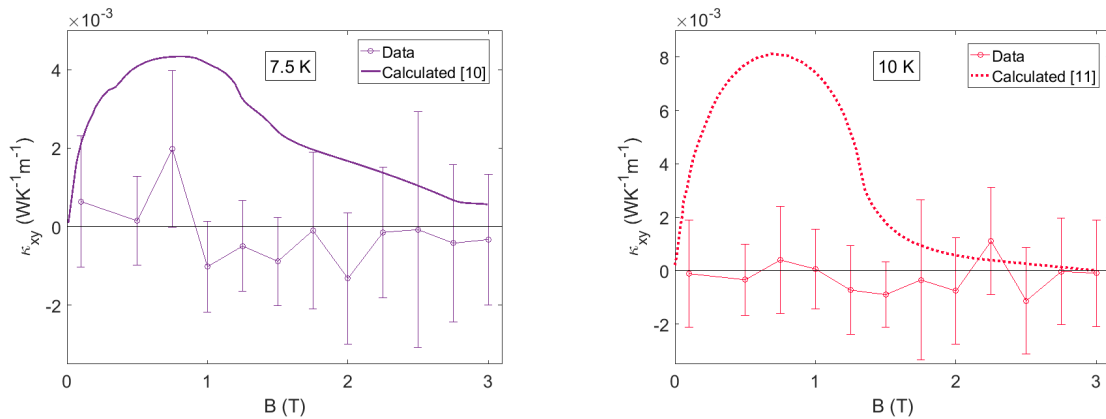


Fig. 4. Transverse thermal conductivity data for $\text{SrCu}_2(\text{BO}_3)_2$, plotted as a function of field in order to compare against calculated curves from [10] and [11]. Errorbars account for standard statistical error, as well as errors in calibration and estimated positions of the thermal contacts.

As acknowledged in both the studies which predict a thermal Hall effect in $\text{SrCu}_2(\text{BO}_3)_2$, there is only a narrow temperature window in which it might be experimentally feasible to detect the signal. At too low a temperature, the thermal occupation of the triplon bands (and therefore triplon density) is insufficient to produce a perceptible thermal current. The calculated filling fraction has been shown to drop off exponentially with decreasing temperature, and is already as low as $\sim 0.2\%$ at ~ 5 K [11]. Likewise, inelastic neutron studies have shown that the intensity of single triplet excitations effectively disappears above 13 K [15], implying that triplons cannot act as thermal carriers at higher temperatures. This provides some justification for the temperature range under investigation, but the

question still remains why there should be no measurable signal.

The accuracy with which the model from McClarty et al [10] is able reproduce the experimentally determined triplon band structure suggests that there is nothing considerable which has been overlooked. Similarly, the same method of calculating a thermal Hall signal has been applied successfully in previous studies of magnonic systems [9, 22], and should therefore be applicable here. Instead, in $\text{SrCu}_2(\text{BO}_3)_2$, it is possible that triplon-triplon interactions play a more significant role than anticipated. These interactions would act to disrupt those (ideally dissipationless) triplon edge states which underpin the thermal Hall effect, and thus reduce the magnitude of any signal. Triplon-triplon interactions will necessarily depend on the thermal occupation of the triplon bands; consequently, future studies might seek to improve the experimental resolution at lower temperatures.

To conclude, we have attempted to verify the theoretical prediction of a thermal Hall effect of triplons in the material $\text{SrCu}_2(\text{BO}_3)_2$, however, within the experimental resolution, no such signal has been observed. This is potentially due to triplon-triplon interactions having more of an inhibitory effect than anticipated. We wish to gratefully acknowledge funding from the UK EPSRC, grant number EP/P013686/1.

Appendix

A.1 Alternative Material - $\text{Lu}_2\text{V}_2\text{O}_7$

In order to verify the experimental procedure, a similar setup was used to investigate a second material - $\text{Lu}_2\text{V}_2\text{O}_7$. This is an insulating ferromagnet with a pyrochlore structure, which has previously been shown to exhibit a thermal Hall effect of a similar magnitude to that anticipated in $\text{SrCu}_2(\text{BO}_3)_2$, albeit with a maximum at a slightly higher temperature [24]. In order to account for this, the thermometers were replaced with CX-1070. Aside from this, the remainder of the experimental setup was identical.

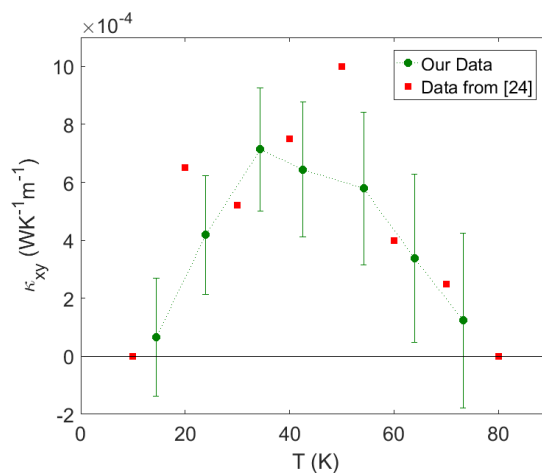


Fig. A.1. Transverse thermal conductivity data for $\text{Lu}_2\text{V}_2\text{O}_7$, plotted as a function of temperature and taken at a constant field of 0.5 T. Green circles represent data from this investigation, red squares are data taken from [24]. As with the $\text{SrCu}_2(\text{BO}_3)_2$ data, those errorbars shown account for standard statistical error, as well as errors in calibration and estimated positions of the thermal contacts.

Shown in Figure A.1 is the transverse conductivity data for $\text{Lu}_2\text{V}_2\text{O}_7$. The data has been taken at just a single field (0.5 T), as this was a test of the experimental method rather than a full investigation.

As expected, there is a clear peak at around 40 K, implying that the same setup should be capable of detecting a similar signal in $\text{SrCu}_2(\text{BO}_3)_2$.

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