

2014

Calorimetric Determination of the Angular Dependent Phase Diagram of an $S = 1/2$ Heisenberg Triangular-lattice Antiferromagnet

Scott T. Hannahs

National High Magnetic Field Laboratory

Nathanael Alexander Fortune

Smith College, nfortune@smith.edu

J.H. Park

National High Magnetic Field Laboratory

Y. Takano

University of Florida

T. Ono

Osaka Prefecture University

See next page for additional authors

Follow this and additional works at: https://scholarworks.smith.edu/phy_facpubs

Part of the [Physics Commons](#)

Recommended Citation

Hannahs, Scott T.; Fortune, Nathanael Alexander; Park, J.H.; Takano, Y.; Ono, T.; and Tanaka, H., "Calorimetric Determination of the Angular Dependent Phase Diagram of an $S = 1/2$ Heisenberg Triangular-lattice Antiferromagnet" (2014). Physics: Faculty Publications, Smith College, Northampton, MA.

https://scholarworks.smith.edu/phy_facpubs/26

This Article has been accepted for inclusion in Physics: Faculty Publications by an authorized administrator of Smith ScholarWorks. For more information, please contact scholarworks@smith.edu

Authors

Scott T. Hannahs, Nathanael Alexander Fortune, J.H. Park, Y. Takano, T. Ono, and H. Tanaka

OPEN ACCESS

Calorimetric determination of the angular dependent phase diagram of an $S=1/2$ Heisenberg triangular-lattice antiferromagnet

To cite this article: S T Hannahs *et al* 2014 *J. Phys.: Conf. Ser.* **568** 042010

View the [article online](#) for updates and enhancements.

Related content

- [Magnetic-field-induced Heisenberg to XY crossover in a quasi-2D quantum antiferromagnet](#)
N A Fortune, S T Hannahs, C P Landee *et al.*
- [Field-induced quantum phase transitions in the spin-1/2 triangular-lattice antiferromagnet \$\text{Cs}_2\text{CuBr}_4\$](#)
N A Fortune, S T Hannahs, Y Takano *et al.*
- [Top-loading small-sample calorimeters for measurements as a function of magnetic field angle](#)
N A Fortune and S T Hannahs

Calorimetric determination of the angular dependent phase diagram of an $S=1/2$ Heisenberg triangular-lattice antiferromagnet

S. T. Hannahs¹, N. A. Fortune², J.-H. Park¹, Y. Takano³, T. Ono⁴
and H. Tanaka⁵

¹National High Magnetic Field Laboratory, Tallahassee, FL, United States of America

²Department of Physics, Smith College, Northampton, MA, United States of America

³Department of Physics, University of Florida, Gainesville, FL, United States of America

⁴Department of Physics, Osaka Prefecture University, Osaka, Japan

⁵Department of Physics, Tokyo Institute of Technology, Meguro-ku, Tokyo, Japan

E-mail: sth@magnet.fsu.edu

Abstract. An antiferromagnetic system on a 2-D triangular lattice leads to geometric topological frustration. This ideal system has been the subject of theoretical investigations. One experimental realization of this system is the compound Cs_2CuCl_4 . Various magnetization, heat capacity, neutron scattering and NMR studies have identified several magnetic transitions when the magnetic field is applied along one of the three principal axes. The current work investigates the evolution of these phases at intermediate angles as the crystal is rotated relative to the magnetic field. These phases were investigated using a novel rotating calorimeter allowing complete coverage of the experimental parameter space. New magnetic phases only existing at intermediate angles have been found.

1. Introduction

Geometric frustration for an antiferromagnetic system has long been studied on a 2-D triangular lattice as the simplest realization of the phenomenon. Geometric frustration even with a simple basic interaction leads to a highly degenerate ground state. Application of a magnetic field reveals the highly complex phase structure Gekht1996. If a Dzyaloshinskii-Moriya interaction is present, different phases dominate when the magnetic field is applied along different crystal lattice directions. The 2-D $S = \frac{1}{2}$ Heisenberg antiferromagnet on a triangular lattice with nearest neighbor interactions is an excellent system to investigate this phenomenon.

The ideal triangular lattice would require that $J = J'$ where J is the antiferromagnetic spin coupling along the base of the triangular lattice and J' is the coupling along the diagonal bonds. In compound Cs_2CuCl_4 the coupling constants are unequal ($J'/J \simeq 0.33$), and the adjacent layers are very weakly coupled ($\approx 0.045J$) [2]. The weak exchange energies $J \simeq 4$ K allow the phase diagram to be accessed at moderate magnetic field. It has been shown that in different orientations the phase diagram is sensitive to very small interactions introducing entirely new phases [3].

The alternating layers of Cs ensure weak coupling between adjacent layers as shown in Figure 1. The most natural start of investigation is to apply a magnetic field along the a



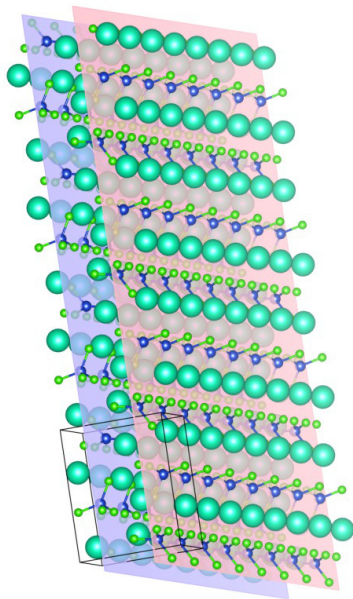


Figure 1. Cs_2CuCl_4 lattice showing inequivalent layered planes (red and blue). Cs atoms dark green and Cu atoms dark blue.

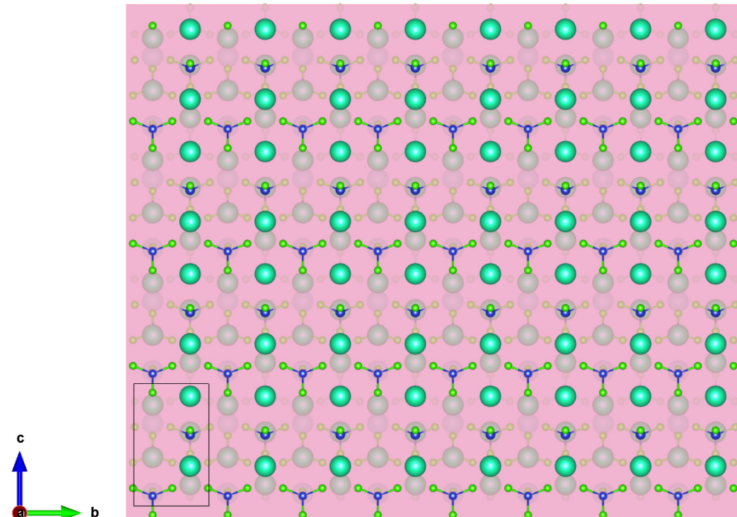


Figure 2. Cs_2CuCl_4 lattice viewed along the a axis showing triangular coordination of Cu atoms (dark blue).

axis perpendicular to the triangular lattice planes. This leads to a very simple phase diagram as derived by Coldea *et al* [4] and shown in Figure 3. The present work re-examines this phase diagram at the temperature profile marked by the red line.

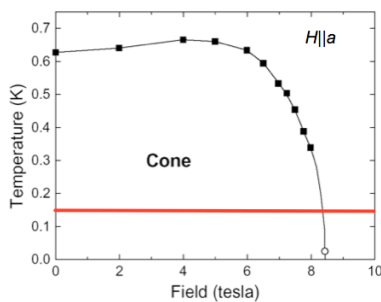


Figure 3. Phase diagram for Cs_2CuCl_4 with the magnetic field applied along the a axis. The cone phase persists and evolves at higher field to the saturated paramagnetic state.

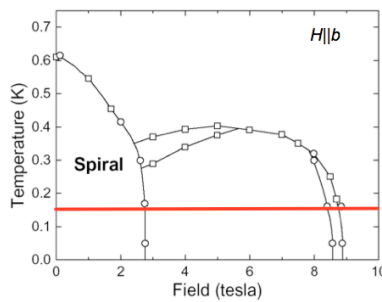


Figure 4. Phase diagram for Cs_2CuCl_4 with the magnetic field applied along the b axis showing more complex behavior with the spiral state and other regions of stability.

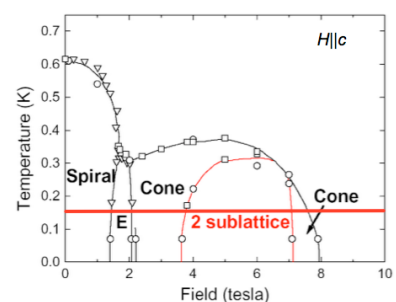


Figure 5. Phase diagram for Cs_2CuCl_4 with the magnetic field applied along the c axis. A complex phase diagram where the spiral state ends at even lower field.

Further work by Tokiwa *et al* [5] extended the phase diagram to fields applied along the b axis (along the triangle bases) and c axis. This reveals a much richer phase diagram with several additional phases identified by NMR measurements [6]. The b axis and c axis are similar – aside from additional phases when the magnetic field is applied along the c axis – but on

their own provide no indication how the cone state of the a axis evolves to the spiral states of the b and c axes. The current work utilizes a unique low temperature rotatable calorimeter [7] using magnetocaloric effects to map the evolution of these phases as the magnetic field is rotated between the principal axes of the lattice.

2. Results

As the magnetic field is swept while holding the sample at constant temperature in the calorimeter, the transitions between phases will involve either a peak in heat capacity or a latent heat depending on the nature of the transition. Our very sensitive determination of phase boundaries can detect small (< 1 mK) changes in temperature as the field is swept. The Cs_2CuCl_4 sample of approximately 1 mg mass was thermally linked to a silver platform in a vacuum cell. The platform was regulated at a constant temperature above the base temperature of the dilution refrigerator. This temperature was regulated independent of field by correcting the magnetic field dependence of the resistance using a previously developed algorithm [8]. Small variations in the sample temperature as the field was swept give the magnitude of the magnetocaloric effect.

The phase diagram was determined at 175 mK for all angles as the calorimeter was rotated from the field along the c axis (most complex) to the a axis (simplest phase diagram). The data are shown in Figure 6, along with the phase identifications as used by Coldea *et al* [2] and Yoshida *et al* [6] as determined by NMR.

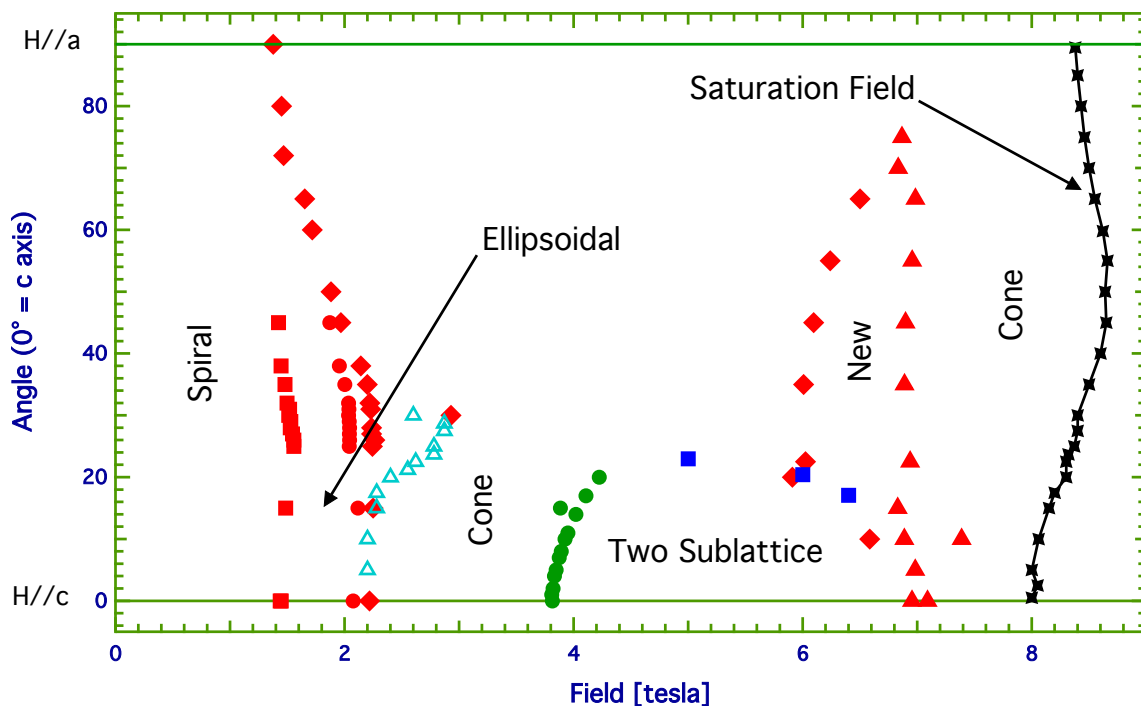


Figure 6. Phase diagram of Cs_2CuCl_4 as the sample angle to the magnetic field is changed. The black x symbols show the saturation field. The black x symbols and the open light blue triangles were measured by torque magnetometry at a temperature of 21 mK [9].

The dome between 3.8 and 7 tesla up to 20° becomes very difficult to resolve in the region where the phase boundary becomes parallel to the abscissa in the figure. In this region the

ordinary magnetocaloric effect becomes very weak. In this case we make use of the gyro-magnetocaloric effect [10], where holding the temperature and magnetic field constant the sample is rotated with respect to the field while recording small changes in temperature. Clearly defined transitions are resolved using this technique. The blue square symbols show transitions determined in this manner.

It is known that Cs_2CuCl_4 has a spiral ground state due to the Dzyaloshinskii-Moriya interactions [2]. We find that the high and low field cone phases merge as the magnetic field vector approaches the a axis above 80° and that these phases must be identical. At angles above 20° where the two-sublattice dome closes a new phase appears that is stable from 20° to 80° and disappears just before the field becomes parallel with the a axis. The narrow ellipsoidal region separating the spiral and cone phases collapses at high angle and disappears resulting in the simple phase diagram along the a axis. Finally note that the phase boundary separating the spiral phase from the cone phase extends continuously to the a axis as an addition to the previous phase diagram shown in Figure 3.

If the sample is rotated from the many transitions of the c axis into the b axis there are similar regions identified but with a different angular dependence. This angular dependence of the phase diagram is shown in Figure 7.

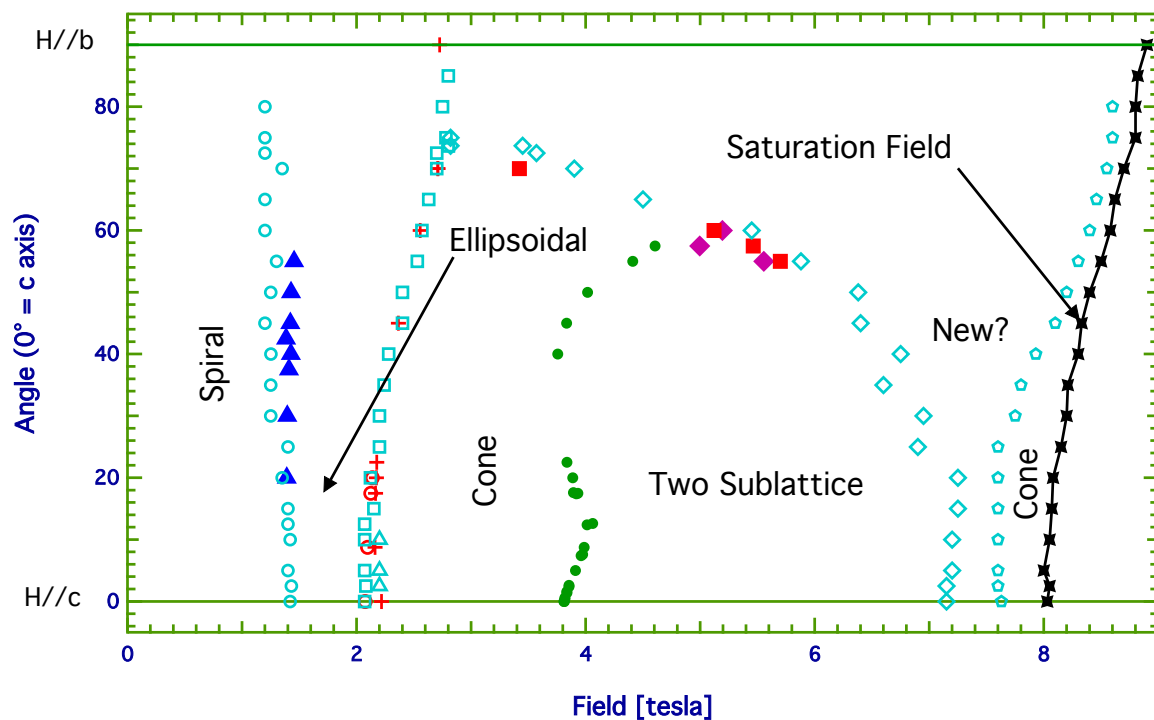


Figure 7. Phase diagram of Cs_2CuCl_4 as the sample angle to the magnetic field is changed. The black x symbols show the saturation field. The black x symbols and the open light blue symbols were measured by torque magnetometry at a temperature of 21 mK [9].

In this case the ellipsoidal region extends to the b axis; however, the transitions between this phase and the spiral phase becomes very weak as the phases merge. The two-sublattice phase extends further from the c axis but eventually closes and disappears at angles higher than 60° . In this phase diagram, the lower-field cone phase also disappears at high angles and does not merge with the higher-field phase which survives at angles up to 90° . The exact nature of the

new high field phase has not been determined by these bulk thermodynamic measurements.

3. Conclusions

We have determined the phase diagram of Cs_2CuCl_4 at low temperature as a function of magnetic field and angle. The phases identified by magnetocaloric and gyro-magnetocaloric effects largely confirm previous work along the three principal crystallographic axes. However, the previous identification of phases along these axes must be modified to account for the tracking of these phase boundaries as a function of angle. The extension of the spiral-cone boundary to the a axis and the appearance of a new intermediate angle phase in this material show that the rich and complex behaviors of this spin $\frac{1}{2}$ triangular-lattice Heisenberg antiferromagnet need further investigation and modeling. Extension of this work to higher temperatures would confirm and map the intermediate angle behavior on the previously determined field-temperature phase diagrams.

Acknowledgments

The authors would like to acknowledge experimental support from the National High Magnetic Field Lab; in particular Scott Maier and Jonathan Billings provided technical assistance. The National High Magnetic Field Lab is supported by National Science Foundation, Cooperative Agreement No. DMR-1157490, the State of Florida, and the U.S. Department of Energy.

References

- [1] Gekht R S and Bondarenko I N, 1997 "Triangular antiferromagnets with a layered structure in a uniform field", *Journal of Experimental and Theoretical Physics* **84** 345.
- [2] Coldea R, Tennant D A, Habicht K, Smeibidl P, Wolters C and Tylczynski Z, 2002, "Direct measurement of the spin Hamiltonian and observation of condensation of magnons in the 2D frustrated quantum magnet Cs_2CuCl_4 " *Phys. Rev. Lett.* **88** 137203.
- [3] Starykh O A, Katsura H and Balents L, 2010 "Extreme sensitivity of a frustrated quantum magnet: Cs_2CuCl_4 ", *Phys. Rev. B* **82** 014421.
- [4] Coldea R, Tennant D A, Tsvelik A M and Tylczynski Z 2001 "Experimental realization of a 2D fractional quantum spin liquid", *Phys. Rev. Lett.* **86** 1335.
- [5] Tokiwa Y, Radu T, Coldea R, Wilhelm H, Tylczynski Z and Steglich F 2006 "Magnetic phase transitions in the two-dimensional frustrated quantum antiferromagnet Cs_2CuCl_4 ", *Phys. Rev. B* **73** 134414.
- [6] Yoshida M, Suzuki T, Waki T and Takigawa M, 2008 Poster presentation at "International Conference on Highly Frustrated Magnetism" 7-12 September 2008 Braunschweig, Germany, Unpublished.
- [7] Hannahs S T and Fortune N A 2003 "Heat capacity cell for angular measurements in high magnetic fields", Proc. of the 23rd International Conference on Low Temperature Physics (LT23), Aug 20-27, 2002 Hiroshima, Japan, *Physica B - Condensed Matter* **329** 1586-1587.
- [8] Fortune N, Gossett G, Peabody L, Lehe K, Uji S and Aoki H 2000 "High magnetic field corrections to resistance thermometers for low temperature calorimetry", *Rev. Sci. Instrum.* **71** 3825-3830.
- [9] Takano Y, Park J.-H., Ono T. and Tanaka H. 2014 Unpublished.
- [10] Nikitin S A, Skokov K P, Koshkid'ko Y S, Pastushenkov Y G and Ivanova T I 2010 "Giant rotating magnetocaloric effect in the region of spin-reorientation transition in the NdCo_5 single crystal", *Phys. Rev. Lett.* **105** 137205.