

# Controlling frustrated magnetism on the kagome lattice by uniaxial-strain tuning

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It is predicted that strongly interacting spins on a frustrated lattice may lead to a quantum disordered ground state or even form a quantum spin liquid with exotic low-energy excitations. However, a thorough tuning of the frustration strength, separating its effects from those of disorder and other factors, is pending. Here we break the symmetry of a kagome-lattice compound in a controlled manner by applying *in situ* uniaxial stress. The transition temperature of  $\text{Y}_2\text{Cu}_5(\text{OH})_{14}\text{OCl}_6$  is linearly enhanced with strain,  $\Delta T_N/T_N \approx 10\%$  upon in-plane compression of order 1%, providing clear evidence for a release of frustration and its pivotal role for magnetic order. Our comprehensive  $^1\text{H}$  NMR results suggest a  $\tilde{Q}^2 = (1/3 \times 1/3)$  state under unstrained conditions and further reveal an incomplete antiferromagnetic transition with fluctuating moments in this strongly frustrated system.

Even after two decades of intense scrutiny, quantum spin liquids (QSL) remain an elusive state of matter [1–4]. Apart from growing evidence for the importance of disorder [5–15], geometrical frustration is considered decisive to suppress magnetic order in presence of strong antiferromagnetic (AFM) exchange interactions. The vast majority of QSL candidates are found in quasi two-dimensional correlated electron systems with triangular [8, 16–21], honeycomb [22] or kagome [23, 24] lattices. Herbertsmithite,  $\text{ZnCu}_2(\text{OH})_6\text{Cl}_2$ , is an archetype realization of the latter symmetry [24] and has been intensely studied over the last two decades [5, 24–31] – not least due to the exciting proposal of exotic superconductivity and Dirac bands in a doped kagome lattice [32]. Although the latter scenario could not be realized so far [33], many related compounds substituting Zn by other bi- or trivalent cations have been synthesized by now [23]. Among those,  $\text{Y}_2\text{Cu}_5(\text{OH})_{14}\text{OCl}_6$  [34] (denoted as Y-kapellasite) and  $\text{YCu}_3(\text{OH})_6\text{Cl}_2$  [35] crystallize in the closely-related kapellasite structure (see Fig. 1) and exhibit AFM order at temperatures  $T_N \ll J/k_B \approx 10^2$  K.

So far, most attempts to modify the frustration strength focused on chemical substitution in order to arrange the valence electrons in the above mentioned patterns. While commonly physical pressure is applied to tune electronic interactions, e.g. towards metal-insulator transitions, hydrostatic compression does not directly affect the lattice symmetry, unless it triggers a structural transition. Recent developments in piezoelectric uniaxial strain applications at cryogenic temperatures [36, 37] now provide us the opportunity to modify the degree of geometrical frustration in a controlled manner.

Here, we take full advantage of uniaxial strain to directly tune magnetic order in Y-kapellasite single crystals. We characterize the magnetic properties by  $^1\text{H}$  nuclear magnetic resonance (NMR) in a temperature range 1.5–200 K and reveal strong spin correlations for  $T < 30$  K. We find AFM below  $T_N = 2.2$  K that is consistent with the proposed  $\tilde{Q}^2 = (1/3 \times 1/3)$  order [38]. By

applying uniaxial strain of order 1% we tune the exchange interactions and frustration strength *in situ* triggering a pronounced increase of  $T_N$  linear with strain.

In the two Y analogs of herbertsmithite (kapellasite), the additional charge upon substitution of  $\text{Zn}^{2+}$  is compensated, resulting in Mott insulators with a charge-transfer gap of 3 eV ( $U \approx 8$  eV [39]). In the case of  $\text{YCu}_3(\text{OH})_6\text{Cl}_2$  this leads to an unstable crystal structure – similar to  $\text{GaCu}_3(\text{OH})_6\text{Cl}_2$  [33] it can be only synthesized as powder [35, 40] – while large  $\text{Y}_2\text{Cu}_5(\text{OH})_{14}\text{OCl}_6$  single crystals with slightly distorted kagome layers (structure shown in Fig. 1) can be grown by hydrothermal methods [34]. Both compounds exhibit AFM order at temperatures much lower than  $\Theta_{\text{Cu-O}} \approx 100$  K; magnetization, specific heat,  $\mu$ -SR and neutron diffraction experiments yield  $T_N = 2.2$  K for Y-kapellasite [34, 40], related with the observation of THz magnons [41], and  $T_N = 15$  K for  $\text{YCu}_3(\text{OH})_6\text{Cl}_2$  [35, 40, 42, 43]. Despite the apparent absence of a QSL state down to  $T \rightarrow 0$ , distorted kagome lattices [44–47] came into focus recently due to magnetoelastic coupling and the realization of

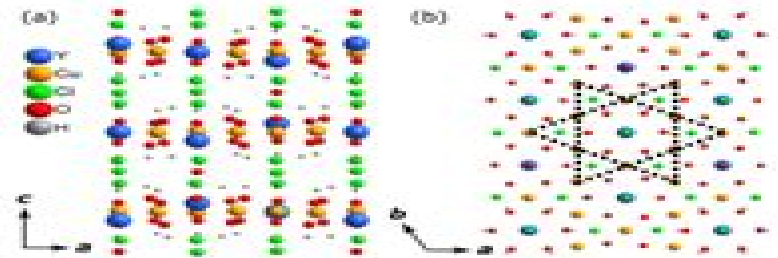


FIG. 1. Crystal structure of  $\text{Y}_2\text{Cu}_5(\text{OH})_{14}\text{OCl}_6$  (Y-kapellasite). (a)  $\text{Cu}^{2+}$  atoms (orange) are arranged in layers parallel to the  $ab$ -plane. (b) Within the plane they form a  $S = 1/2$  kagome lattice indicated by black dotted lines.

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1. Understanding the eBook Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
  - The Rise of Digital Reading Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
  - Advantages of eBooks Over Traditional Books
2. Identifying Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
  - Exploring Different Genres
  - Considering Fiction vs. Non-Fiction
  - Determining Your Reading Goals
3. Choosing the Right eBook Platform
  - Popular eBook Platforms

- Features to Look for in an Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
- User-Friendly Interface
- 4. Exploring eBook Recommendations from Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
  - Personalized Recommendations
  - Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977 User Reviews and Ratings
  - Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977 and Bestseller Lists
- 5. Accessing Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977 Free and Paid eBooks
  - Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977 Public Domain eBooks
  - Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977 eBook Subscription Services
  - Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977 Budget-Friendly Options
- 6. Navigating Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977 eBook Formats
  - ePub, PDF, MOBI, and More
  - Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977 Compatibility with Devices
  - Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977 Enhanced eBook Features
- 7. Enhancing Your Reading Experience
  - Adjustable Fonts and Text Sizes of Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
  - Highlighting and Note-Taking Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
  - Interactive Elements Proceedings Of The International Conference On Lattice Dynamics Paris September 59

1977

8. Staying Engaged with Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
  - Joining Online Reading Communities
  - Participating in Virtual Book Clubs
  - Following Authors and Publishers Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
9. Balancing eBooks and Physical Books Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
  - Benefits of a Digital Library
  - Creating a Diverse Reading Collection Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
10. Overcoming Reading Challenges
  - Dealing with Digital Eye Strain
  - Minimizing Distractions
  - Managing Screen Time
11. Cultivating a Reading Routine Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
  - Setting Reading Goals Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
  - Carving Out Dedicated Reading Time
12. Sourcing Reliable Information of Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
  - Fact-Checking eBook Content of Proceedings Of The International Conference On Lattice Dynamics Paris September 59 1977
  - Distinguishing Credible Sources
13. Promoting Lifelong Learning
  - Utilizing eBooks for Skill Development
  - Exploring Educational eBooks
14. Embracing eBook Trends
  - Integration of Multimedia Elements

- Interactive and Gamified eBooks

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