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 Y₃Cu₉(OH)₁₈OC₁₈₈ is linearly enhanced with strain, ΔTN/TN ≈ 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

¹H NMR results suggest a Q = (1/3 × 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, ZnCu₃(OH)₆Cl₂, 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 bivalent cations have been synthesized by now [23]. Among those, Y₃Cu₉(OH)₁₈OC₁₈₈ [34] (denoted as Y-kapellasite) and YCu₁₈(OH)₆Cl₃ [35] crystallize in the closely-related kapellasite structure (see Fig. 1) and exhibit AFM order at temperatures TN ≪ J/kB ≈ 10² 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 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 TN = 2.2 K that is consistent with the proposed Q = (1/3 × 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 TN linear with strain.

In the two Y analogs of herbertsmithite (kapellasite), the additional charge upon substitution of Zn²⁺ is compensated, resulting in Mott insulators with a charge-transfer gap of 3 eV (U ≈ 8 eV [39]). In the case of YCu₁₈(OH)₆Cl₃ this leads to an unstable crystal structure — similar to GaCu₁₈(OH)₆Cl₃ [33] it can be only synthesized as powder [35, 40] — while large Y₃Cu₉(OH)₁₈OC₁₈₈ 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 ΘCW ≈ 10 K; magnetization, specific heat, µ-SR and neutron diffraction experiments yield TN = 2.2 K for Y-kapellasite [34, 40], related with the observation of THz magnons [41], and TN = 15 K for YCu₁₈(OH)₆Cl₃ [35, 40, 42, 43]. Despite the apparent absence of a QSL state down to T → 0, distorted kagome lattices [44–47] came into focus recently due to magnetoelectric coupling and the realization of

FIG. 1. Crystal structure of Y₃Cu₉(OH)₁₈OC₁₈₈ (Y-kapellasite). (a) Cu²⁺ 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.
non-trivial ground states such as pinwheel [48, 49] and \( Q = (1/3 \times 1/3) \) order [38].

Prior to application of strain we carried out a thorough ambient-pressure NMR characterization of Y-kapellasite. The magnetic field was aligned parallel to the \( Cu^{2+} \) chains of the kagome network, i.e. perpendicular to the crystallographic \( a \)-axis; in the following, this in-plane field configuration is denoted as \( B \parallel ab \). Fig. 2a presents the temperature evolution of the \(^1\)H spectra \((B_0 = 0.9789 \ T, \nu_0 = 41.68 \ MHz)\), yielding two peaks separated by 30–40kHz above 4 K. At lower temperatures, the line broadening exceeds the experimental bandwidth such that the intensity (probed by integrating the spectral weight \( SW \)) deviates from the standard \( T^{-1} \) dependence. To resolve the entire peak structure, we performed magnetic field sweeps at constant frequency \( \nu_0 = 41.68 \ MHz \), revealing a pronounced splitting into three peaks below \( T_N = 2.2 \ K \). The separation between the two outer peaks \((B_1, B_2)\) increases to lower temperatures (inset) while the central peak remains at \( B_0 \). The grey line indicates the spectrum at 6 K, which follows the frequency scale at the top.

(c) The false-color plot of the data from panel (b) illustrates the shifting of \( B_1 \) and \( B_2 \) as well as the strong broadening. The horizontal bar corresponds to a frequency of \( \Delta \nu = \Delta B/\gamma = 1 \ MHz \).

While our observation of a line splitting evidences AFM order setting in at \( T_N \), the ratio among the peak intensities provides insight into the details of the spin structure. Upon \( Q = (1/3 \times 1/3) \) order predicted for a distorted kagome lattice [38], along a \( Cu^{2+} \) chain four successive AFM bonds are satisfied followed by two unsatisfied bonds. We point out that \(^1\)H nuclei are ideal probes of the local field at a bond as they are attached to oxygen atoms and, hence, are sitting between two neighboring \( Cu^{2+} \) sites, as sketched in Fig. 3a. If the electron spins are antiparallel, their local fields cancel out yielding a proton resonance at \( B_0 \); for parallel alignment, their local fields add up to (subtract from) the external field yielding the NMR line at \( B_1 \) \((B_2)\). Our Gaussian fits in Fig. 3b indeed reveal an intensity ratio between \( B_0; B_1; B_2 \) sites of 4.09:3.07:1 that is close to 4:1:1 [50]. Overall, the spectral features of our NMR measurements are consistent with \( Q = (1/3 \times 1/3) \) order in Y-kapellasite [38] and motivate more thorough assessment of this model.

Apart from the splitting, the NMR linewidth (Fig. 3c) is subject to severe broadening. The full width at half maximum (FWHM) increases beyond the homogeneous linewidth \((\pi T_2)^{-1} \approx 35 \ kHz\) already for \( T < 30 \ K \), indicating the onset of antiferromagnetic fluctuations, followed by an abrupt broadening at \( T_N \). Note, while the FWHM increases by two orders of magnitude across the studied temperature range, the spin-spin relaxation rate merely varies by a factor two. The two-peak structure of the spectrum \((T > 4 \ K)\) and the absolute values of \( T_2 \) are caused by proton-proton dipolar coupling since hyperfine coupling to the spin-1/2 \( Cu^{2+} \) sites is weak.
(a) $Q = (1/3 \times 1/3)$ AFM order predicted for a distorted kagome lattice [38]. Colored circles indicate location of $^1$H nuclei in the crystal lattice of Y-kapellasite (cf. Fig. 1). (b) The NMR intensity (violet) at 1.7 K was fitted with three Gaussian peaks corresponding to the $^1$H positions (see color in (a)); their intensity scales as 4:0.93:1.07 ($B_0$; $B_1$; $B_2$). The spin-lattice relaxation rate (orange diamonds) was measured at different fields: $T_1^{-1}$ of the side peaks ($B_1$, $B_2$) is $\approx 50\%$ smaller than for the central peak at $B_0 = 0.9789$ T. (c) The full width at half maximum (FWHM), extracted for the $^1$H NMR peaks $H_1$ and $H_2$ ($T > T_N$; see Fig. 2a) as well as for the three lines $B_0$, $B_1$ and $B_2$ emerging below $T_N$ (see Fig. 2b,c), exceeds the homogeneous linewidth below 30 K and shows a steep increase at $T_N$. Note the different units of the vertical scales.

Hence, we assign the pronounced line broadening below 30 K to magnetic correlations in Y-kapellasite.

Aside from $T_2$ effects, the spin-lattice relaxation rate $T_1^{-1}$ is highly susceptible to the emerging magnetism. Above 10 K the $^1$H relaxation rate of Y-kapellasite, plotted in Fig. 4, is of similar magnitude compared to ZnCu$_3$(OH)$_6$Cl$_2$ [51]. However, herbertsmithite exhibits a different temperature dependence with a maximum around 90 K and steadily decreasing $T_1^{-1}$ upon cooling below that (magenta dotted line in Fig. 4). Here, we observe a positive slope at high temperatures, likely related to lattice dynamics, while below 30 K the relaxation rate increases upon cooling due to AFM fluctuations. Down to 4 K we do not observe pronounced field dependence or anisotropy (measurements for $B \parallel c$ shown in inset of Fig. 4) in $T_1^{-1}$.

The onset of magnetic order yields a sharp peak at the transition temperature $T_N = 2.2$ K. Increasing the magnetic field to 2 T and 3.7 T decreases the peak values of $T_1^{-1}$ and broadens the maximum, in accord with specific heat results [34]. As AFM sets in, relaxation rapidly decreases upon further cooling. Throughout, we determined the temperature dependence of $T_1^{-1}$ at $B_0$. At 1.7 K we also measured $T_1^{-1}$ at the two outer peaks $B_1$ and $B_2$ (Fig. 3b) yielding a 50% smaller value compared to $B_0$. The faster relaxation at the central peak indicates fluctuating spins – possibly a small portion that has evaded (or is not involved in) static AFM order. In this regard, NMR measurements at lower temperatures are highly desired, especially at $T \ll 1$ K.

To summarize the findings so far, the NMR properties are consistent with AFM order of $\mathbf{Q} = (1/3 \times 1/3)$ symmetry [38]. Moreover, Y-kapellasite features coexistence of fluctuating and static moments, in line with recent $\mu$-SR results [40]. The spin-lattice relaxation rate is highly susceptible to the onset of magnetic order and exhibits a sharp peak at the transition, making it a sensitive probe of the transition temperature. Having identified its unusual magnetic ground state, in the following we apply in-plane uniaxial stress to Y-kapellasite single crystals. Through distorting the kagome lattice by compression along the Cu$^{2+}$ chains, as sketched in Fig. 5d, we directly modify the anisotropy of the transfer integrals $t'/t$ and,
hence, the exchange interactions $J$, $J'$ and their degree of geometrical frustration.

In Fig. 5 we trace the change in transition temperature through $T_N^{-1}$ measurements at $B_0 = 1.81$ T ($B \parallel a$) upon applying uniaxial stress parallel to the Cu$^{2+}$ chains. There is a clear enhancement of $T_N$ with approximately linear dependence on the applied strain, which agrees with calculations of Heisenberg AFM ordering on a distorted kagome lattice [52]. Our results on two samples consistently reveal an increase of transition temperature by almost 10% for compressive strain of order 1% parallel to the kagome layers. How does this compare with the modifications expected for hydrostatic pressure of similar size? For comparison, we consider that the superexchange is proportional to $t^4/(\Delta^2U)$ and $t \propto r^{-4}$, hence 1% reduction of Cu-Cu distance should result in an increase of 16%. However, this is a large overestimate, because in real materials the crystal lattice adapts mostly by changing bond angles rather than bond length [53]. Thus, we conclude that the major effect arises from releasing frustration of the kagome lattice.

Let us assess the observed strain-tuning effects in the context of other materials where the structure impacts the magnetic properties. In ZnCu$_3$(OH)$_6$Cl$_2$, indications of DM interaction [24, 29] and even symmetry breaking [54, 55] have been reported, likely related to magnetoelastic coupling [56]. Hydrostatic pressure yields an enhancement of magnetic order [57], but also a field-induced spin freezing was reported [58]. The compound studied here shows structural similarities to the 'pin-wheel' kagome structure of Rb$_2$Cu$_3$SnF$_{12}$ [48, 59] and a Barlowite polymorph [49]. To that end, Y-kapellasite is likely frustrated in a different way than Herbertsmithite or Kapellasite, which both exhibit an undistorted kagome lattice – at the expense of severe Zn/Cu antisite disorder [6]. Finally, an indication for a structural involvement in the magnetic degrees of freedom is the onset of spin fluctuations below a recently discovered structural anomaly at 32 K [60]. An intriguing scenario could be that this is the onset of nematic order below a crossover at $T^* \gg T_N$ predicted for a distorted kagome lattice [52]. Extending our present strain-tuning experiments on Y-kapellasite to higher temperatures would enable direct scrutiny of this issue and thus remains a desideratum for future work.

To conclude, we performed comprehensive $^1$H NMR investigations on Y-kapellasite as a function of temperature, magnetic field and uniaxial stress. Under unstrained conditions we find spectral evidence for AFM order of $\mathcal{Q} = (1/3 \times 1/3)$ type below $T_N = 2.2$ K in this distorted kagome compound. Clear signatures for AFM are also seen in the linewidth and spin-lattice relaxation rate, which are dominated by proton dynamics at elevated temperatures while magnetic correlations become dominant below 30 K. Based on this characterization, we applied in situ uniaxial stress parallel to the kagome layers, resulting in a linear increase of $T_N$ with strain, in accord with theoretical predictions [52]. Through a controlled release of frustration, our findings evidence its crucial importance for correlated quantum magnets and spin liquids, opening the door for similar studies in triangular, honeycomb and other kagome materials. The pioneering strain-tuning approach presented here is ideally suited to resolve the magnetic ground states of existing materials, and to discover novel exotic spin phases beyond state-of-the-art theoretical frameworks [2, 4].

**METHODS**

NMR experiments were performed using variable temperature $^4$He cryostats and superconducting magnets. At $T \geq 4$ K magnetic field and frequency were fixed according to the $^1$H resonance condition $\nu_B = \gamma B$, with $\gamma = 42.5774$ MHz/T, as the proton NMR spec-
trum was resolved within the experimental bandwidth. Due to excessive line broadening below 4 K, the NMR intensity started to drop below the expected $T^{-1}$ behavior, as illustrated by blue circles in the inset of Fig. 2a that correspond to the integrated spectral weight (SW) of the NMR spectrum. To that end, at $T < 4$ K the $^1$H spectra were acquired by sweeping the external field in dense intervals around $B_0 = 0.9789$ T. As illustrated by the black diamonds in the inset of Fig. 2a, the entire NMR intensity was recovered in these field-sweep measurements: in the inspected field range the total SW of the $^1$H resonance lines up with the $T^{-1}$ behavior extrapolated from $T \geq 4$ K. Zero-strain NMR measurements were performed using large single crystals ($3 \times 2 \times 1$ mm$^3$).

The relaxation measurements (Fig. 4) for in-plane (measured at UCLA) and out-of plane (acquired at TU Wien on a different sample) crystal axes show similar $\nu_{1}$ values. The relaxation rate $T_1^{-1}$ was acquired at the central line ($\nu_0$, $B_0$ in Fig. 2b). At $T = 1.70$ K we measured $T_1^{-1}$ at $\nu_0 = 41.68$ MHz also for the two satellite peaks $B_1$ and $B_2$ obtaining values approximately 50% of $T_1^{-1}$ / $B_0 = 0.9789$ T (orange symbols in Fig. 3b).

Uniaxial strain experiments were performed using piezoelectric strain cells (Razorbell) in the same $^4$He cryostat as the zero-strain studies for in-plane magnetic fields. For that, the Y-kapellasite sample (the single crystal shown in Fig. 5a-c was cut in dimensions $2.5 \times 0.45 \times 0.24$ mm$^3$ with the longest dimension parallel to the Cu chains of the kagome layer, i.e. $\epsilon \parallel a$) was glued at room temperature on the strain cell using Stycast and dried for several days. Here, the magnetic field direction was parallel to the $a$-axis, i.e. perpendicular to the in-plane fields in Fig. 2. Still, we observed a similar peak in $T_1^{-1}$ at $T_N$ for both $B \parallel a$ (Fig. 5) and $B \parallel ab$ (Fig. 4), hence our measurements of the relaxation rate were suitable to trace the strain dependence of the AFM transition. The strain voltage was varied at 4 K and successively $T_1^{-1}$ was measured upon cooling through the transition, both for increasing and decreasing strain as indicated by red and orange symbols in Fig. 5d, respectively. The compression was monitored in situ by the built-in capacitive displacement sensor. Similar to previous strain studies [61, 62], the applied strain $\epsilon$ was calculated as the displacement divided by the length of the sample surface that remained free of epoxy – for sample a2 this corresponds to 0.6 mm (Fig. 5a-c). The linear increase of $T_N$ with strain shown in the inset of Fig. 5d lines up for the two samples. Still, we do not exclude that the real compression of the sample is somewhat lower than the present estimate using the built-in capacitive sensor, likely $\epsilon_{\text{real}} \approx 1\%$; also in Sr$_2$RuO$_4$ the strain of the van Hove singularity was first estimated as 0.6% [61, 63] and later corrected to 0.44% by using high-precision stress-strain sensors [64].

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