Chasing the spin gap through the phase diagram of a frustrated Mott insulator

The quest for entangled spin excitations has stimulated intense research on frustrated magnetic systems. For almost two decades, the triangular-lattice Mott insulator $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ has been one of the hottest candidates for a gapless quantum spin liquid with itinerant spinons. Very recently, however, this scenario was overturned as electron-spin-resonance (ESR) studies unveiled a spin gap, calling for reevaluation of the magnetic ground state. Here we achieve a precise mapping of this spin-gapped phase through the Mott transition by ultrahigh-resolution strain tuning. Our transport experiments reveal a reentrance of charge localization below $T^\ast = 6$ K associated with a gap size of 30–50 K. The negative slope of the insulator-metal boundary, $dT^\ast/ dp < 0$, evidences the low-entropy nature of the spin-singlet ground state. By tuning the enigmatic ‘6K anomaly’ through the phase diagram of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, we identify it as the transition to a valence-bond-solid phase, in agreement with previous thermal expansion and magnetic resonance studies. This spin-gapped insulating state persists at $T \rightarrow 0$ until unconventional superconductivity and metallic transport proliferate.

Since Anderson’s notion of resonating valence bonds$^1$, the quest for quantum spin liquids (QSL) has been fueled by the idea that suppressing long-range antiferromagnetic (AFM) order can stabilize itinerant or even topological spin excitations$^2$–$^4$. What is often neglected is that the coupling of magnetic degrees of freedom to the lattice can result in valence-bond-solid (VBS) phases with paired electron spins. Such singlet states occur, for instance, in triangular-lattice organic compounds$^4$–$^8$ and in form of the well-known spin-Peierls (SP) states in quasi 1D systems$^{9,10}$. Also in other frustrated materials the importance of valence-bond phases has been revived$^{11,12}$. Such a VBS scenario is fully consistent with the report of a spin gap in the organic QSL candidate $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, that aroused great attention in the QSL community very recently$^{13,14}$, in agreement with gapped excitations in thermal transport$^{15}$ and a phase transition at 6 K deduced from thermal expansion$^{16}$ and NQR measurements$^{17}$. While this finding rules out a gapless scenario with mobile spinons, that would have explained the finite Sommerfeld coefficient in specific heat results$^{18}$, it leaves open the possibility of gapped QSL phases$^{19,20,21}$.

The tunability of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$—a pressure of 1.3 kbar triggers the transition from a Mott insulator to superconducting (SC) and metallic phases$^{22,23}$, as illustrated in Fig. 1d—provides the unique opportunity to scrutinize the entropy of its magnetic ground state. According to the Clausius-Clapeyron relation, the pressure-tuned metal-insulator transition (MIT) acquires a positive slope $dT_{\text{mit}}/ dp > 0$ in the $T$-$p$ phase diagram between a paramagnetic Mott insulator with large spin entropy and a metal (Fig. 1b), reminiscent of the Pomeranchuk effect in $^3$He$^{28}$. On the other hand, the entropy of AFM and VBS states is smaller than that of the Fermi liquid (FL) metal, yielding negative slopes $dT_{\text{N}}/ dp < 0$ and $dT^\ast/ dp < 0$ of the MIT (Fig. 1a, c), respectively.

Here we utilize the slope of the insulator-metal boundary of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ as a probe of the spin-gapped ground state below $T^\ast = 6$ K, evidencing its low-entropy nature. For that, we map the low-temperature Mott MIT with ultrahigh precision by measuring dc transport upon a combination of biaxial compression and tensile strain. We find a reentrance of insulating behavior setting in below $T^\ast$, which is successively suppressed as
metallicity proliferates. Through comparison with previously reported structural\textsuperscript{28,39} and magnetic\textsuperscript{40} properties, our present results clearly identify the ‘6k anomaly’ as the transition to a PBS phase with \(dT/dp<0\) (Fig. 1c) and, thus, we complete the phase diagram of \(\kappa\)-(BEDT-TTF)\(_2\)Cu\(_2\)(CN)\(_3\). Moreover, our transport data exhibit a gap size of 30–50 K for the spin-singlet state, which coincides with the unexpectedly large critical field of 60 T\(^1\).

**Results**

Among the QSL candidates, \(\kappa\)-(BEDT-TTF)\(_2\)Cu\(_2\)(CN)\(_3\) has been studied most intensely as it exhibits not only frustrated AFM exchange interactions (\(J<250 \text{ K}\))\textsuperscript{22,30}, but also a paradigmatic Mott transition\textsuperscript{22,29,30}. This layered triangular-lattice compound with \(t'/t\) close to unity\textsuperscript{33,32} shows no AFM order\textsuperscript{26} and, at the same time, it is one of the best solid-state realizations of the single-band Hubbard model\textsuperscript{29}. As such, it provides experimental access to check the predictions of dynamical mean-field theory; while the Mott transition is first-order type below the critical endpoint \(T_{\text{crit}}\), a quantum-critical crossover at the quantum Widom line (QWL) occurs at \(T>T_{\text{crit}}\)\textsuperscript{29,34,31,32}. Moreover, applying 1–2 kbar pressure allows to probe unconventional SC\textsuperscript{32,35} as well as charge transport in Fermi liquids and bad metals\textsuperscript{30}. By combining the most recent findings in \(\kappa\)-(BEDT-TTF)\(_2\)Cu\(_2\)(CN)\(_3\)\textsuperscript{28,30,35} and compounds with different degree of frustration\textsuperscript{28,29,30,34,31}, in Fig. 1d we present the state-of-the-art phase diagram of dimerized organic Mott systems as a function of \(T\), electronic correlations \(U/W\) and geometrical frustration \(t'/t\).

In this study, we explore the phase space extremely close to the MIT, which requires fine tuning of the correlation strength with a resolution equivalent to 10\(^{-3}\) kbar. While such high precision cannot be reached in conventional oil pressure cells\textsuperscript{25,33}, also gas pressure experiments are limited at low temperatures due to solidification of helium\textsuperscript{27,30}. To that end, we utilize strain transmitted through a substrate—a method previously applied in doping-tuned experiments\textsuperscript{39}. As sketched in Fig. 2a, a single-crystalline \(\kappa\)-(BEDT-TTF)\(_2\)Cu\(_2\)(CN)\(_3\) filament of 140 nm thickness is placed on a flexible polyethylene substrate at ambient conditions. Differential thermal expansion between sample and substrate (see Methods) causes an in-plane biaxial compression \(\varepsilon_{\text{biaxial}}\) equivalent to 1–2 kbar hydrostatic pressure, which is sufficient to push the sample across the Mott MIT (Fig. 2b). Our dc transport results in Fig. 2c yield metallic properties and an onset of SC below \(T_c=4 \text{ K}\) (\(\varepsilon_{\text{c}}=0\), see inset). Note, uniaxial strain experiments on \(\kappa\)-(BEDT-TTF)\(_2\)Cu\(_2\)(CN)\(_3\) revealed a considerable enhancement of \(T_c\) and a broadened SC dome\textsuperscript{40} than for isotropic compression. Here, biaxial strain does not affect the in-plane anisotropy and \(T_c\) is comparable to hydrostatic pressure experiments\textsuperscript{32,27}.

While differential thermal contraction tunes the sample across the Mott MIT, it does not allow to vary the correlation strength in arbitrary steps. In order to reach back into the Mott state in a quasi-continuous manner, we perform band-tuning via tensile strain \(\varepsilon_{\text{c}}<0\) along the crystallographic c-axis. As indicated on the bottom of Fig. 2a, we control the correlation strength by varying the bending radius of the substrate, which yields a negative pressure of less than a kilobar. As such, uniaxial strain is significantly weaker than the overall pressure applied (\(\varepsilon_{\text{u}}<|\varepsilon_{\text{biaxial}}+\varepsilon_{\text{c}}|\)), hence we do not expect major effects from a change in frustration \(t'/t\) certainly less pronounced than in ref. 40. The blue shaded region in Fig. 2b indicates the phase space covered in our experiments with a fixed \(\varepsilon_{\text{biaxial}}<0\) and varying tensile strain between \(\varepsilon_{\text{c}}=0\) and 0.32%. The resulting transport curves in Fig. 2c exhibit a textbook Mott MIT: the resistivity maxima at the Brinkman-Rice temperature \(T_{\text{br}}\) are pushed to lower temperatures as correlations increase\textsuperscript{25,33}. In addition, in the range between \(\varepsilon_{\text{c}}=0.17\%\) and 0.25% we observe a distinct upturn of resistance upon cooling well
In ambient pressure transport studies\cite{29}, the regime $T < 10 \text{K}$ remained inaccessible due to the extremely large resistance; here, in the vicinity of the MIT $R(T)$ is small enough to be measured. Following its observation in thermodynamic and magnetic probes\cite{22}, in our present work we identify the ‘6k anomaly’ in the metallic state of $\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3$ for the first time in charge transport as a local maximum of $d\ln R/dT$ at $6 \text{K}$. Even more importantly, we obtain a quantitative estimate of the associated energy gap $E_g \approx 10 \text{K}$ for $\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3$.

In Fig. 4a, we compare the structural changes at the ‘6k anomaly’ in $\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3$ with the VBS transition at $T^* = 24 \text{K}$ in EtMe$_2$P[Pd(dmit)$_2$]$_2$. Thermal expansion experiments by Manna et al.\cite{30} revealed very similar anisotropic distortions within the conducting layers at $T^*$. In both quasi 2D organic compounds, as sketched in Fig. 4a, the crystal contracts along the $c$-axis and expands along the other in-plane direction upon cooling. Also other probes observed pronounced anomalies at 6K due to modifications of the crystal structure and possible symmetry breaking, which are typical fingerprints of a VBS transition.

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Altogether, these findings provide solid evidence for anisotropic structural changes at the transition to a singlet state with a gap in the spectrum of spin excitations. In addition, $T^\ast - 6\, \text{K}$ at $p = 0$ directly interpolates to the reentrance of the MIT in our strain-dependent transport data, which is mapped in the false-color plot of $d\ln R/dT$ vs $1/T$ in Fig. 4d. We find that the resulting phase diagram prominently displays $d^2T/ dp^2 < 0$ as sketched in Fig. 1c, analogous to the VBS Mott insulator $\text{EtMe}_3\text{P}[(\text{dmit})_2\text{Pd}]/\text{Cu}_2(\text{CN})_3$ with similar thermal expansion anomalies (Fig. 4a, b). In light of these overwhelming experimental similarities, we conclude that at ambient pressure the enigmatic ‘6K anomaly’ in $\kappa$-$(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3$ is the transition from a paramagnetic Mott insulator to a spin-gapped VBS state upon cooling, as observed here for 0.17% (BEDT-TTF)$_2$Cu$_2$(CN)$_3$ dissolved. We applied a current of 8 mA overnight and obtained tiny thin crystals of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$. The crystals were pipetted to 2-propanol, and were manipulated with a tip of hair and put on the substrate. After the substrate was taken out from 2-propanol and dried, the thin crystal tightly adhered to the substrate.

**Discussion**

Since the first reports of the ‘6K anomaly’ and the absence of magnetic order$^{1,2}$, there have been different interpretations of the experimental results acquired over two decades in independent groups$^{3-5,14,16}$. Here, we put together now all these pieces and obtain a comprehensive understanding of the low-temperature physics of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$. Our present ultrahigh-resolution tuning of the ‘6K anomaly’ through the Mott MIT delivers the missing part required to establish it as a true phase transition to a low-entropy spin-singlet ground state with distorted crystal structure. In view of the very shape of the phase diagram presented here, the magnetic, thermal transport, thermodynamic and structural properties of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ are consistent with a spin-gapped VBS state, essentially identical to that reported in other layered organic Mott systems$^{35}$. While chiral QSL states with a spin gap$^{32,33}$ cannot be ruled out at present, it is unclear whether the distinct lattice effects of the ‘6K anomaly’$^\ast$ are compatible with these theoretical models. In absence of a ‘smoking-gun’ experiment deciding between the two possibilities, the principle of Occam’s Razor$^{34}$ suggests to favor not the exotic (chiral QSL) but rather the established scenario (VBS), not least as the latter has already been observed in related triangular-lattice systems$^{3-5}$.

As such, we point out that the spin-gapped low-entropy phase associated with the ‘6K anomaly’ is not intrinsic to pure Mott insulators, but rather results from magneto-structural instabilities. Likely, the elastic energy involved in the lattice distortion at $T^\ast$ contributes to the unexpectedly large energy gap and critical field, exceeding the spin gap deduced from ESR data$^4$ and $k_B T^\ast/\mu_B = 9 \, \text{T} \, \text{for}^3$. Previous works have suggested the involvement of charge degrees of freedom$^{20,41}$, but a sizeable charge disproportionation has been ruled out by vibrational spectroscopy$^{16}$. The recent observation of reentrant insulating behavior in the related QSL candidate $\kappa$-(BEDT-TTF)$_2$Ag$_2$(CN)$_3$ possibly provides another test bed to investigate these issues$^{41}$.

Finally, we suggest that, in absence of the VBS, the metallic state would persist much deeper into the insulating region of the phase diagram—in Fig. 2b $T_F$ and $T_{\text{INS}}$ extrapolate to zero well below $p = 1 \, \text{kbar}$$^{13,35}$. To that end, elucidating the genuine Mott MIT in the limit $T \rightarrow 0$ requires to suppress $T^\ast$ in $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, potentially by varying $T$/$t$ through uniaxial strain (Fig. 1d) or by applying high magnetic fields $B = 50$–100 T. To conclude, our findings highlight the importance to gain understanding and control of magneto-elastic coupling in frustrated spin systems, in particular in other QSL candidates.

**Methods**

**Sample preparation and application of uniaxial bending strain**

We prepared polyethylene terephthalate (PET) substrates (Teflon® FT7, Toyobo Film Solutions Limited) and patterned 18-nm-thick Au electrodes using photo-lithography. Thin single crystals of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ were synthesized with electrolysis of a 1,1,2-trichloroethane [10% (v/v) ethanol] solution (50 ml), in which BEDT-TTF (20 mg), KCN (60 mg), CuCN (40 mg), and 18-crown-6 (100 mg) are dissolved. We applied a current of 8 nA overnight and obtained tiny thin crystals of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$. The crystals were pipetted to 2-propanol, and were manipulated with a tip of hair and put on the substrate. After the substrate was taken out from 2-propanol and dried, the thin crystal tightly adhered to the substrate.
As described in previous work, cooling yields differential thermal expansion between sample and substrate: the length change is \( \pm 1\% \) between 0 K and 300 K for \( \kappa \)-(BEDT-TTF)\(_2\)Cu\(_2\)(CN)\(_3\), compared to \( >1\% \) for PET. As a result, the sample is subject to biaxial compression of order 1–2 kbar, deduced from the transport results in Fig. 2 in comparison to previously published resistivity data. We applied uniaxial bending strains by pushing the back of the substrate with a nanopositioner (ANPzS1, attocube systems) as shown in Fig. 2a. Assuming that the bent substrate is an arc of a circle, the strain \( \varepsilon \) is estimated as \( \varepsilon = \frac{4\pi(t^2 + 4x^2)}{4t} \) using the small angle approximation, where \( x \) is the displacement of the nanopositioner, \( t = 177 \mu \text{m} \) and \( l = 12 \text{ mm} \) are the thickness and length of the substrate, respectively. The uniaxial tensile strain was applied at 100 K in descending order from 0% to 3.32%. The cooling rate of the sample was 0.5 K/min. The thickness of the studied \( \kappa \)-(BEDT-TTF)\(_2\)Cu\(_2\)(CN)\(_3\) crystal (140 nm) was measured \textit{a posteriori} using a step profiler. Throughout the manuscript we follow the convention that a reduction of the sample volume (upon compression stress or pressure) corresponds to negative strain \( \varepsilon < 0 \), whereas an expansion of the sample size (upon tensile stress or ‘negative’ pressure) yields \( \varepsilon > 0 \). By comparing our results to transport measurements under hydrostatic pressure, we estimate that the total applied strain \( \varepsilon_{\text{applied}} \) corresponds to a hydrostatic pressure \( p = 1.55 \text{ kbar} \) for \( \varepsilon = 0 \), and \( p = 1.05 \text{ kbar} \) for \( \varepsilon \approx 0.32\% \), yielding a conversion factor \( \Delta \varepsilon / \Delta p = 0.6 \% / \text{ kbar} \). The range (1.05–1.35 kbar) covered by tuning of \( \varepsilon \) in Figs. 2c and 4d is indicated by the blue rectangle in Fig. 2b.

Data analysis
The QWL in Fig. 2b was taken from ref. 28 whereas \( T_{\text{NRM}} \), \( T_c \), and \( T_N \) are from ref. 35 and the insulator-metal boundary below 20 K (cyan) was determined from the resistivity data in ref. 27. The transport gap of \( \kappa \)-(BEDT-TTF)\(_2\)Cu\(_2\)(CN)\(_3\) at ambient pressure in Fig. 3 (dashed black line) was taken from Fig. S2 in the Supplementary Information of ref. 28; the gray dotted line is a rigid vertical shift of the ambient pressure data by \(-130 \text{ K} \). \( \Delta \varepsilon_{\text{max}} \) was determined for the outer \( ^6\text{Li} \) nuclei using a chemical shift \( \chi < 117 \), as described in ref. 17. In the same panel, the ESR susceptibility \( \chi \) for magnetic field perpendicular to the layers \( B // \text{||} \) \( \text{||} \) (similar to the Knight shift data) was taken from Fig. 2a in ref. 16. Both \( \chi \) and \( K_{\text{outer}} \) were normalized to their value at 10 K.

At elevated temperatures the resistivity maxima indicate \( T_{\text{NRM}} \). At lower temperatures in the phase coexistence regime \( T \approx T_{\text{coexistence}} \approx 15 \text{ K} \), strictly speaking, the resistivity maxima indicate percolation of metallic regions. We assign the sharp jumps in the resistance data in Fig. 2c, also seen as narrow peaks between 10–15 K in Fig. 3, to an artefact of the sample mounting. As such, the apparent strain dependence is naturally related to the volume change of the sample that interferes with the applied strain. A micro-crack will open or close for a particular differential thermal expansion between sample and substrate, reached at a different temperature for different strain conditions upon changing \( \varepsilon \).

Data availability
The authors declare that the data supporting the findings of this study are available within the paper. Further information can be provided by A.P. and Y.K. upon request.

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Additional information

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