Unambiguous Experimental Verification of Linear-in-Temperature Spinon Thermal Conductivity in an Antiferromagnetic Heisenberg Chain

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The everlasting interest in spin chains is mostly rooted in the fact that they generally allow for comparisons between theory and experiment with remarkable accuracy, especially for exactly solvable models. A notable example is the spin-$\frac{1}{2}$ antiferromagnetic Heisenberg chain (AFHC), which can be well described by the Tomonaga-Luttinger liquid theory and exhibits fractionalized spinon excitations with distinct thermodynamic and spectroscopic experimental signatures consistent with theoretical predictions. A missing piece, however, is the lack of a comprehensive understanding of the spinon heat transport in AFHC systems, due to difficulties in its experimental evaluation against the backdrop of other heat carriers and complex scattering processes. Here we address this situation by performing ultralow-temperature thermal conductivity measurements on a nearly ideal spin-$\frac{1}{2}$ AFHC system copper benzoate $Cu(C_6H_5COO)_2 \cdot 3H_2O$, whose field-dependent spin excitation gap enables a reliable extraction of the spinon thermal conductivity κ_s at zero field. κ_s was found to exhibit a linear temperature dependence $\kappa_s \sim T$ at low temperatures, with κ_s/T as large as 1.70 mW cm^{-1} K^{-2}, followed by a precipitate decline below $\sim$0.3 K. The observed $\kappa_s \sim T$ clarifies the discrepancies between various spin chain systems and serves as a benchmark for one-dimensional spinon heat transport in the low-temperature limit. The abrupt loss of κ_s with no corresponding anomaly in the specific heat is discussed in the context of many-body localization.

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At its core, the enigmatic quantum spin liquid (QSL) phase is characterized by long-range quantum entanglement, fractionalized excitations, and emergent gauge s tructures. One often resorts to the characterization of fractionalized excitations (spinons, etc.) for the identification and classification of QSLs of various types [1–5]. These efforts, however, have yet to establish a consensus for two- and three-dimensional (2D and 3D) systems. An outstanding puzzle is that, despite the purported fingerprints of spinons in thermodynamic and spectroscopic probes, unambiguous observation of spinon heat transport is still absent [6–24]. Resolving the discrepancies between different probes necessitates a comprehensive understanding of the impacts of various perturbations to the model Hamiltonians, as they often lead to magnetic ground states competing with a QSL and, concomitantly, substantial change in the excitation spectrum. Such an understanding is rarely available in 2D and 3D systems [1–5].

The conceptual simplicity of one-dimensional (1D) systems has proven useful in the mitigation of the issues mentioned above [25,26]. For instance, the spin-$\frac{1}{2}$ antiferromagnetic Heisenberg chain (AFHC) is a prototypical strongly interacting quantum many-body system, featuring well-defined spinon excitations [Fig. 1(a)] [27,28]. Indeed, the spinon continuum [Fig. 1(b)]—a spectroscopic hallmark of spinons—has been observed, consistent with analytical results at a quantitative level, in several AFHC systems, including copper benzoate $Cu(C_6H_5COO)_2 \cdot 3H_2O$ [29–33]. However, even for such "simple" cases, previous efforts in establishing the characteristic spinon heat transport remain inconclusive. In the low-temperature limit, the contribution from itinerant spinons to the thermal conductivity κ_s was proposed to follow either a linear or quadratic temperature dependence, i.e., $\kappa_s \sim T$ or $\kappa_s \sim T^2$ [34–38]. The direct comparison with experimental data to distinguish between the two has been hindered by the large uncertainty in the subtraction of the phonon thermal conductivity κ_{ph}, mostly a consequence of various scattering processes present at the relatively high measurement temperatures [39–44]. As most quantum magnets of interest are electrical insulators, heat transport has become a unique probe of the charge-neutral spin excitations, especially in determining their itinerant vs localized nature [9,22,24,45]. Clarifying the debate regarding the characteristic spinon heat transport constitutes an essential part of the complete theoretical description of AFHC systems and

would provide crucial guidance for the study of QSLs in higher dimensions.

In this Letter, we investigate spinon heat transport by performing ultralow-temperature specific heat and thermal conductivity measurements on high-quality single crystals of copper benzoate. Gapless spinons dominate the excitation spectrum at zero field, as expected from the AFHC model and confirmed by a linear-temperature-dependent specific heat $C_s \sim T$. Under a magnetic field, soft modes develop at incommensurate wave vectors connecting the Zeeman-split spinon Fermi points [46]. However, the field-induced spin excitation gap is beyond the description of the AFHC model [46,47]. Instead, an induced staggered field originating from the staggered g tensor and Dzyaloshinskii-Moriya interaction renders copper benzoate amenable to another integrable model, the quantum sine-Gordon model, featuring solitons, antisolitons, and their bound states, dubbed breathers [46–48]. The well-established field dependence of the spin excitation gap allows for a reliable estimation of $\kappa_{\rm ph}$ which, in turn, enables an unbiased evaluation of κ_s at zero field. κ_s was found to exhibit a linear temperature dependence $\kappa_s \sim T$ at low temperatures, consistent with the ballistic spinon heat transport limited solely by chain impurities or defects and in sharp contrast to the prediction of $\kappa_s \sim T^2$ from a spin-boson picture. Unexpectedly, κ_s decreases abruptly below ~0.3 K before it vanishes at ~0.1 K, while no concomitant anomaly in C_s was observed, hinting at intriguing possibilities regarding many-body localization (MBL).

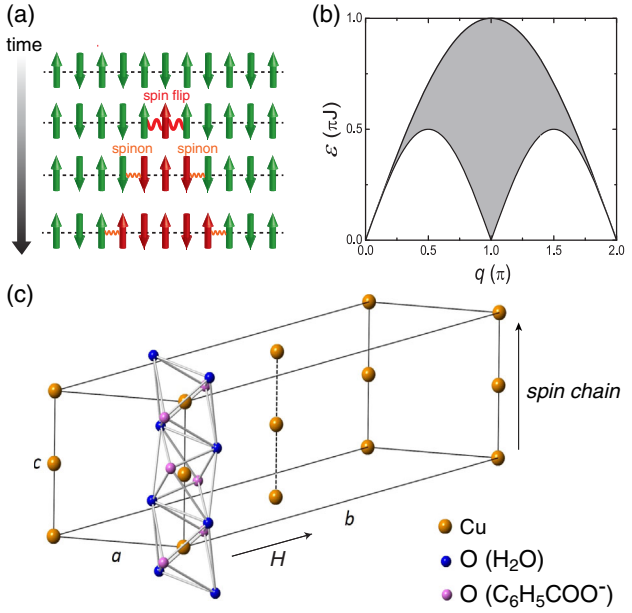

FIG. 1. (a) Schematics showing the generation of spinons in a spin-$\frac{1}{2}$ antiferromagnetic chain. An initial spin flip results in two unsatisfied bonds that can freely propagate, acting as domain walls and dubbed spinons. (b) Typical spectrum for spinons, featuring a continuum with clear boundaries. (c) The crystal structure of copper benzoate $Cu(C_6H_5COO)_2 \cdot 3H_2O$. The Cu^{2+} ions carry spin-$\frac{1}{2}$ and form chains along the c axis. Each spin site locates in the center of a distorted octahedron formed by oxygen ions. The intrachain coupling $J \sim 18$ K and the extremely weak interchain coupling J' ($T_N \sim 0.8$ mK) make copper benzoate a model compound of the spin-$\frac{1}{2}$ antiferromagnetic Heisenberg chain. For all measurements, the external magnetic field was applied along b.

As shown in Fig. 1(c), copper benzoate crystallizes in the monoclinic space group $I2/c$ [29,46]. The Cu^{2+} spin chains run along the c axis, with an antiferromagnetic intrachain exchange coupling $J \sim 18$ K [29,46]. Reflecting the nearly ideal one dimensionality, the antiferromagnetic ordering temperature T_N is as low as 0.8 mK (hence an extremely weak interchain coupling J') [49,50]. The thermal conductivity of copper benzoate single crystals was measured in a dilution refrigerator via a standard four-wire steady-state method with thermal contacts attached to the ac plane using silver paint. The heat current J_Q was applied along the spin chain for three samples, namely $S1$, $S2$, and $S3$, and perpendicular to the chain for $S4$. The magnetic fields were applied along the b axis for all samples in both specific heat and thermal conductivity measurements. We exploit specific procedures to account for heat leak, thermal contact resistance, and other factors that may affect the data quality at ultralow temperatures. With these procedures, the experimental error bar can be as low as $\kappa/T = \pm 0.005$ mW cm^{-1} K^{-2}. More details about the growth, characterization of the samples, and the measurement procedure can be found in Sec. I of the Supplemental Material [51].

The specific heat C is displayed in Fig. 2. The absence of a Schottky anomaly is attributed to the small J combined with the low impurity (defect) concentration (see Sec. IV in the Supplemental Material [51] for more details). At zero field, the data from 0.05 to 3.5 K are fitted to $C = C_s + C_{\rm ph} = \gamma T + \beta T^3$ [solid line in Fig. 2(a)], where C_s and $C_{\rm ph}$ are the contributions from gapless spinons and phonons, respectively. The fitting parameters are $\gamma = 0.291 \pm 0.001$ J mol^{-1} K^{-2} and $\beta = 0.0094 \pm 0.0001$ J mol^{-1} K^{-4}. The T-linear C_s is better illustrated in the inset of Fig. 2(a) as a plateau in the temperature dependence of C_s/T at low temperatures. An estimation based on the Bethe ansatz yields $C_s/T = 0.298$ J/mol K^2 [46,76,77], in excellent agreement with the value of γ obtained from the fitting in Fig. 2(a) and the plateau height in the inset. The magnetic specific heat $C_{\rm mag}$ is plotted for $\mu_0 H = 0$ and 7 T in Fig. 2(b). $C_{\rm mag}(0\ {\rm T})$ is exactly C_s, while $C_{\rm mag}(7\ {\rm T})$ is the specific heat of the gapped solitonlike excitations predicted in the quantum sine-Gordon model [46,47]. By fitting to an empirical formula [solid line in Fig. 2(b), see Sec. II in the Supplemental Material [51] for more details], the spin excitation gap at 7 T is found to be $\Delta \sim 2.2$ K, consistent with previous reports [46,47].

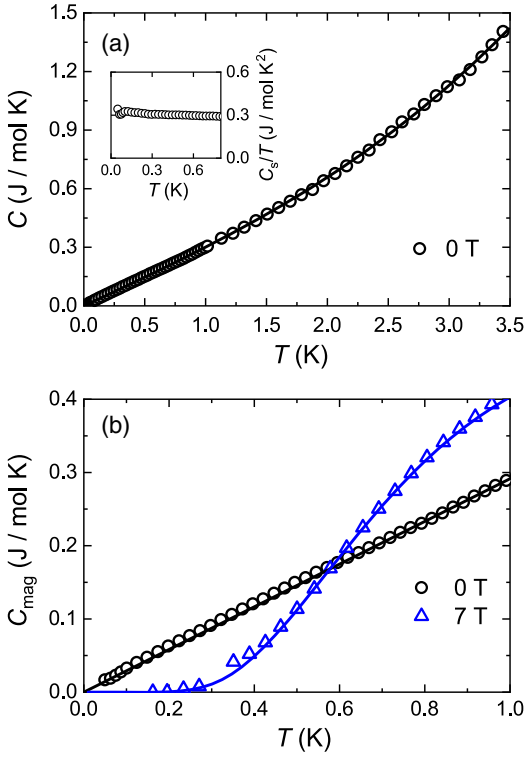

FIG. 2. The specific heat of copper benzoate. (a) The total specific heat C at zero magnetic field. The black solid line is a fit to $C = C_s + C_{ph} = \gamma T + \beta T^3$, where C_s and C_{ph} are the contributions from gapless spinons and phonons, respectively. The linear behavior of C_s is better illustrated as a temperature-independent C_s/T in the inset. (b) Magnetic specific heat C_{mag} at $\mu_0 H = 0$ and 7 T, obtained by subtracting the small C_{ph} determined from the fitting in (a). In the temperature range of our measurement, $C_{mag}(0\ \mathrm{T}) = C_s$ is linear (black solid line), attributed to spinons, while $C_{mag}(7\ \mathrm{T})$ comes from gapped solitonlike excitations. The blue solid line is a fit to extract the spin excitation gap (see text).

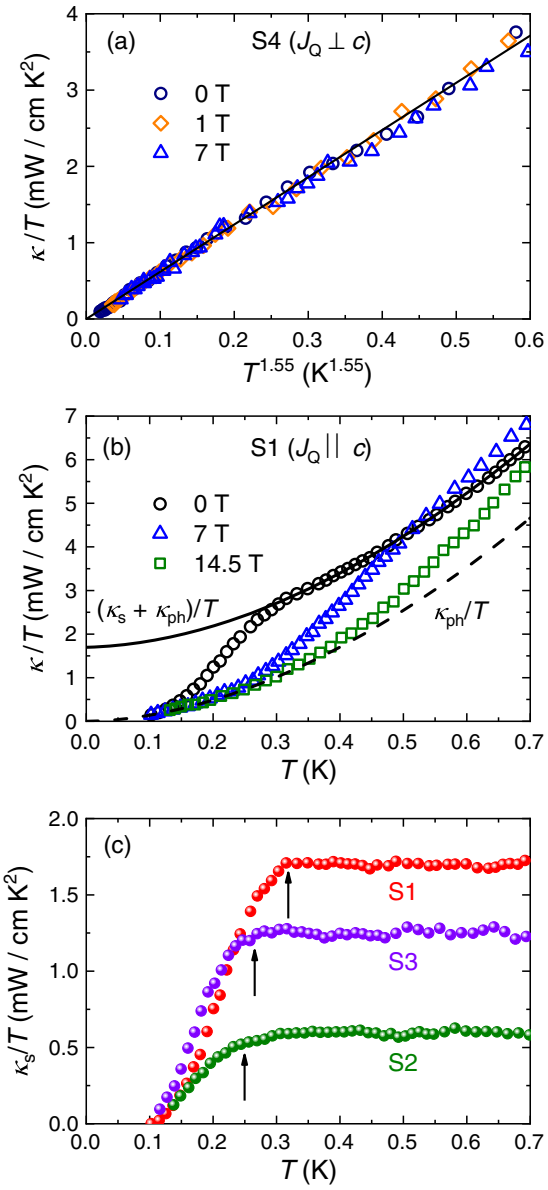

FIG. 3. The thermal conductivity of copper benzoate. (a) The thermal conductivity of $S4$ ($J_Q \perp c$) plotted as κ/T against $T^{1.55}$. A straight line fits well the data under different fields. (b) The thermal conductivity of $S1$ ($J_Q \| c$) plotted as κ/T against T. At 0 T, the data above 0.3 K can be fitted by $\kappa/T = (\kappa_s + \kappa_{ph})/T = a + bT^{\alpha-1}$, where κ_s and κ_{ph} are the contributions from gapless spinons and phonons, respectively. At 14.5 T, κ_{ph}/T obtained from fitting the zero-field data accounts for the data below 0.3 K. (c) The zero-field κ_s/T of various samples $S1$, $S2$, and $S3$, plotted as κ_s/T vs T. The arrows indicate the abrupt drop.

The thermal conductivity κ of various copper benzoate samples is shown in Fig. 3. Generally, similar to the case of specific heat, the thermal conductivity can be decomposed as $\kappa = \kappa_{mag} + \kappa_{ph}$. For the sample $S4$ with a heat current J_Q perpendicular to the chain, the κ data curve under zero and finite fields almost overlap, as seen in Fig. 3(a). This is expected since all spin excitations in copper benzoate are confined within the chains due to the nearly ideal one dimensionality. Therefore, $\kappa_{mag} = 0$ and $\kappa = \kappa_{ph} = (6.19 \pm 0.06)T^{(2.55\pm0.01)}$ for $S4$ under various fields [solid line in Fig. 3(a)]. The unaffected κ_{ph} by magnetic field indicates that, despite the abundant spin excitations with a dynamical crossover from gapless spinons at zero field to gapped solitonlike excitations under finite fields, the scattering of phonons by these spin excitations is negligible to first order in the temperature range measured (see discussions in Sec. V in the Supplemental Material [51]). The phonons are hence in the boundary-scattering limit, where the exponent in the temperature dependence of κ_{ph} deviates from the ideal case of 3 due to specular reflections at sample surfaces [78,79].

The κ of sample $S1$ with J_Q along the chain is displayed in Fig. 3(b). At zero field, spinons and phonons are the only

heat carriers so that $\kappa_{\rm mag} = \kappa_s$ and $\kappa = \kappa_s + \kappa_{\rm ph}$. Assuming $\kappa_s \sim T$—which will be justified later, the zero-field data above ~0.3 K can be well fitted to

$$\kappa = aT + bT^{\alpha}, \tag{1}$$

giving $a = 1.70 \pm 0.04$ mW cm^{-1} K^{-2}, $b = 8.87 \pm 0.04$ mW cm^{-1} K$^{-3.80}$, and $\alpha = 2.80 \pm 0.03$ [solid line in Fig. 3(b)]. Previously, such a large $\kappa_s/T(T \to 0)$ was only initially reported in the 2D organic QSL candidate $EtMe_3Sb[Pd(dmit)_2]_2$ [7], but was not able to be reproduced by other groups later [14,15]. At $\mu_0 H = 7$ and 14.5 T, the heat carriers are the solitonlike excitations and phonons. The spin excitations are only expected to make a contribution when they are thermally activated across the field-induced gap. At 14.5 T, a gap of $\Delta \sim 3.8$ K renders the spin excitations ineffective below ~0.3 K, i.e., $\kappa = \kappa_{\rm ph}$ [46,47]. As demonstrated above for $S4$, $\kappa_{\rm ph}$ is field-independent in copper benzoate. Indeed, for $S1$, the $\kappa_{\rm ph}$ obtained from fitting the zero-field data to Eq. (1), i.e., $\kappa_{\rm ph} = 8.87T^{2.80}$, describes well the 14.5 T data below ~0.3 K [dashed line in Fig. 3(b)]. Therefore, the validity of the assumption of $\kappa_s \sim T$ in Eq. (1) is corroborated by three independent arguments: (i) $\kappa(0\ \mathrm{T})$ is well described by Eq. (1) above ~0.3 K [Fig. 3(b) for $S1$ and Fig. S2(a) in the Supplemental Material [51] for $S2$ and $S3$]; (ii) $\kappa(14.5\ \mathrm{T})$ below ~0.3 K is dominated by the $\kappa_{\rm ph}$ obtained from $\kappa(0\ \mathrm{T})$ [Fig. 3(b) for $S1$]. (iii) Importantly, as shown in Fig. S2(b) in the Supplemental Material [51], for $S1$, $S2$, and $S3$, the fitting of $\kappa(0\ \mathrm{T})$ to Eq. (1) generates a list of values of the coefficient b that is proportional to $\sqrt{S}$, where S is the cross-sectional area of the respective sample. This is the characteristic behavior of $\kappa_{\rm ph}$ in the boundary-scattering limit [52]. These arguments collectively ensure the reliable subtraction of $\kappa_{\rm ph}$ and, consequently, a reliable extraction of κ_s.

The T-linear κ_s is better illustrated as a plateau above ~0.3 K in Fig. 3(c) for three samples. The observed $\kappa_s \sim T$ forms the main finding of this Letter. To our knowledge, *unambiguous* identification of a linear-in-temperature spinon thermal conductivity has not been achieved in previous reports. We elaborate on this below.

The integrability of the AFHC model dictates a divergent κ_s due to a nonzero thermal Drude weight $D_{\rm th}$ [34,35,80,81]. In real materials, the singularity is removed by the scattering of spinons by other spinons, phonons, and impurities. It has been established that, at ultralow temperatures, the former two mechanisms are subdominant compared to the spinon-impurity scattering [38–40,80]. In this regime, $\kappa_s \sim D_{\rm th}\tau_s$, where τ_s is a relaxation time, and $D_{\rm th} \sim v_s T$ [34,35,81]. Only spinons in a small portion of the Brillouin zone matter at low T, rendering the spinon velocity v_s constant [28,41]. Therefore, the T dependence of κ_s is determined by that of τ_s. The observed $\kappa_s \sim T$ points to a T-independent τ_s and spinon mean free path $l_s = v_s\tau_s$, which can be estimated from [40,41]

$$l_s = \frac{3\hbar}{\pi {k_B}^2 n}\frac{\kappa_s}{T}, \tag{2}$$

where $n = 2/ac$ is the number of spin chains per unit area. For $S1$, we obtain a large $l_s \sim 1060$ Å (see Sec. IV in the Supplemental Material [51] for a comparison with other quantum magnets), corresponding to ~330 spin sites along the chain, indicating the low level of impurities (defects) in copper benzoate [41]. Indeed, estimations from l_s and the Curie tail in the magnetic susceptibility χ consistently confirm the low impurity (defect) concentration (see Sec. IV in the Supplemental Material [51] for more details).

The T-independent l_s and linear-in-T κ_s have also been concluded in heat transport studies on the AFHC system Sr_2CuO_3 ($T_N \sim 5$ K) and $SrCuO_2$ ($T_N \sim 2$ K) [42–44], as well as the more ladderlike $CaCu_2O_3$ ($T_N \sim 25$ K) [41]. However, in these cases, the estimation of l_s is only possible for $T >$ ~50 K, due to the large uncertainty in the subtraction of $\kappa_{\rm ph}$ at lower temperatures. Such a temperature is sufficiently low compared to the energy scale of $J \sim 10^3$ K of these materials. However, it is too high relative to the Debye temperature $\Theta_{\rm D} \sim 10^2$ K to safely omit the influence of spinon-phonon scattering and the complicated functional form of $\kappa_{\rm ph}$ itself. In fact, on a plot of l_s against T often found in these studies, there is barely any data point in the T range where one can directly tell whether l_s would decrease with decreasing T or level off eventually [42–44,53]. In contrast, benefiting from the impurity scattering of spinons and boundary scattering of phonons as the only relevant scattering mechanisms, our observation of linear-in-T κ_s and T-independent l_s in copper benzoate places the ballistic heat transport of spinons on solid ground.

On the other hand, within a bosonization framework, $l_s \sim T$ and $\kappa_s \sim T^2$ for the spin-boson excitations [82] were proposed for the low-T limit of the AFHC model [36–38]. This scenario was also argued to agree with the data of Sr_2CuO_3 and $SrCuO_2$ [36–38,43,44]. Again, the comparison with experimental data is only available in a temperature range where spinon-phonon scattering dominates. In yet another AFHC system $Cu(C_4H_4N_2)(NO_3)_2$, $\kappa_s \sim T^2$ was experimentally verified down to ~0.5 K [83]. However, caution must be used when comparing these results with the spin-boson scenario. The theory assumes $J \gg \Theta_{\rm D}$ [36], a criterion clearly met by Sr_2CuO_3 and $SrCuO_2$, but not fulfilled in copper benzoate ($J \sim 18$ K) or $Cu(C_4H_4N_2)(NO_3)_2$ ($J \sim 10$ K). This may partially resolve the discrepancy between the spin-boson scenario and the $\kappa_s \sim T$ observed in copper benzoate, while blurring the connection between the theory and the observed $\kappa_s \sim T^2$ in $Cu(C_4H_4N_2)(NO_3)_2$.

A caveat of the spin-boson scenario is that it deals with the weak impurity limit or, equivalently, $T \gg T_{\mathrm{KF}}$, where T_{KF} is the Kane-Fisher scale, the lower bound of the $\kappa_s \sim T^2$ regime [36–38]. The observed $\kappa_s \sim T$ in $CaCu_2O_3$ was attributed to a defect level too high for the spin-boson scenario to be applicable [41]. Below T_{KF}, scattering by the chain-breaking defects might supposedly lead to a T-independent l_s [41,43,44]. Alternatively, it was argued that, similar to the case of weak impurities, $l_s \sim T$ would be retained even in the presence of strong impurities by tunneling through the impurity potential [83]. Because of the finite interchain coupling, spinons can also bypass the strong impurities by hopping to the neighboring chains. The bypassing mechanism was argued to change the mean free path from a T-linear one as expected from a spin-boson scenario to the observed T-independent behavior in the spin-one Haldane chain $Ni(C_2H_8N_2)_2NO_2(ClO_4)$ [54]. Considering the extremely small J' in copper benzoate, this mechanism seems irrelevant to the observed $\kappa_s \sim T$.

We observed a rapid drop of κ_s below ~0.3 K [Figs. 3(b) and 3(c)]. In the Appendix and Sec. V in the Supplemental Material [51], we discuss its connection to MBL and the nontrivial implications for magnetic heat transport in general.

In summary, by measuring the ultralow-temperature thermal conductivity of a prototypical spin-$\frac{1}{2}$ antiferromagnetic Heisenberg chain copper benzoate, we managed to unambiguously identify a linear-in-temperature spinon contribution, characteristic of ballistic spinon transport constrained by impurities (defects). Such spinon transport is practically the intrinsic one as the divergent transport dictated by conservation laws always gets cut off by the inevitable impurities (defects) in real materials. The data presented here offer an opportunity to make an unbiased comparison with various theoretical scenarios for spinon heat transport in 1D quantum magnets. As the existence of highly mobile fractionalized excitations in higher dimensions remains elusive, the establishment of intrinsic spinon transport in 1D sets the cornerstone for searching for the enigmatic QSL phase in higher dimensions.

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Appendix: The low-temperature drop of spinon thermal conductivity.—The most natural explanation for the rapid drop of κ_s below ~0.3 K is the localization of spinons. Localization of spin excitations in (quasi-)1D systems has rarely been reported, except for the spin ladder compound $(C_5H_{12}N)_2CuBr_4$ and the Ising chain material $CoNb_2O_6$ [45,55]. However, there the localization results in a negligible κ_s in the whole temperature range measured, in contrast to the metal-insulator-transitionlike behavior observed here in copper benzoate. The textbook example of localization in the presence of disorder is the Anderson localization. The fate of such localization under interactions and the existence of a finite-temperature localization transition have been debated for decades [84,85]. It was exactly the theoretical and numerical study on paradigmatic spin chain models (e.g., a random-field XXZ chain) that offered definitive answers to these questions and promoted the research field of MBL [56–58]. The study on MBL keeps broadening the scope of quantum statistical mechanics and bears a potential in quantum information technologies. Experimentally, however, the study of MBL is mostly limited to synthetic systems [56–58]. The few studies in solid-state materials include a novel correlation metric to detect MBL showcased for the nuclear spins in $Ca_5(PO_4)_3F$ [86] and a dramatic drop of electrical conductivity in the field-induced insulating phase of the superconducting amorphous InO below ~0.1 K [59]. The latter was argued to present a remarkable experimental realization of a finite-temperature MBL transition in solid-state materials [59–62]. It is tempting to associate the observed loss of κ_s in copper benzoate with the MBL transition of spinons. However, such a connection is not straightforward, mainly due to an intrinsic contradiction: while an isolated system is required for MBL, the measurement of κ_s requires finite spinon-phonon coupling. The reduced strength of such coupling may mask the $\kappa_s \sim T$ behavior and engender an apparent loss of κ_s, reminiscent of the loss of electron thermal conductivity in certain cuprate superconductors [63,87]. However, as demonstrated in Sec. V in the Supplemental Material [51], the absence of a corresponding anomaly in the C_s of copper benzoate cannot be readily integrated into this scenario, and the loss of κ_s observed here may have nontrivial implications for magnetic heat transport, in general, and warrants further investigations.

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