Evidence of one-dimensional magnetic heat transport in the triangular-lattice antiferromagnet Cs$_2$CuCl$_4$

E. Schulze, S. Arsenijevic, L. Opherden, A. N. Ponomaryov, J. Wosnitza, T. Ono, H. Tanaka, and S. A. Zvyagin

$^1$Dresden High Magnetic Field Laboratory (HLD-EMFL) and Würzburg-Dresden Cluster of Excellence ct.qmat, Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany
$^2$Institute of Solid State and Materials Physics, TU Dresden, 01062 Dresden, Germany
$^3$Department of Physical Science, Osaka Prefecture University, Osaka 599-8531, Japan
$^4$Department of Physics, Tokyo Institute of Technology, Tokyo 152-8551, Japan

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We report on low-temperature heat-transport properties of the spin-1/2 triangular-lattice antiferromagnet Cs$_2$CuCl$_4$. Broad maxima in the thermal conductivity along the three principal axes, observed at about 5 K, are interpreted in terms of the Debye model, including the phonon Umklapp scattering. For thermal transport along the $b$ axis, we observed a pronounced field-dependent anomaly, close to the transition into the three-dimensional long-range-ordered state. No such anomalies were found for the transport along the $a$ and $c$ directions. We argue that this anisotropic behavior is related to an additional heat-transport channel through magnetic excitations, that can best propagate along the direction of the largest exchange interaction. Besides, peculiarities of the heat transport of Cs$_2$CuCl$_4$ in magnetic fields up to the saturation field and above are discussed.

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Frustrated magnets are known to be an excellent playground to test fundamental concepts of condensed matter physics and quantum mechanics [1–3]. Spin-1/2 Heisenberg antiferromagnets (AFs) on triangular lattices have attracted particular attention, representing an important class of low-dimensional (low-D) frustrated magnets and allowing one to probe effects of the geometrical frustration, magnetic order, and quantum fluctuations in strongly correlated spin systems. Particularly, this interest was stimulated by the idea of the “resonating valence bond” (RVB) ground state for an AF system of spins on a triangular layer lattice [4]. This quantum-disordered ground state was proposed to be a 2D fluid of resonating spin-singlet pairs, with the elementary excitation spectrum formed by fractionalized mobile quasiparticles, spinons. Since that time, searching for experimental realizations of the 2D quantum spin liquid appears to be one of the central topics in quantum physics.

Among others, the triangular-lattice AF Cs$_2$CuCl$_4$ has drawn a particular great deal of attention. Magnetic Cu$^{2+}$ ions in Cs$_2$CuCl$_4$ form a quasi-2D lattice with the exchange coupling $J$ along the $b$ axis (regarded as the chain direction) and $J'$ along the zigzag bonds in the $bc$ plane (Fig. 1). Its spin Hamiltonian reads

$$
\mathcal{H}_{2d} = \sum_i J \vec{s}_i \cdot \vec{s}_{i+1} + \sum_k J' \vec{s}_k \cdot \vec{s}_{k+1} + \mathcal{H}_\delta, \quad (1)
$$

where $\vec{s}_i$ are the spins along and $\vec{s}_k$ between the chains (along the zigzag bonds), and $\mathcal{H}_\delta$ represents various possible, usually small, contributions (such as interlayer and the Dzyaloshinskii-Moriya (DM) interactions). High-field electron spin resonance (ESR) measurements [5] revealed $J/k_B = 4.7$ K and $J'/k_B = 1.4$ K ($k_B$ is the Boltzmann constant), yielding the ratio $J'/J \approx 0.3$, which is in good agreement with parameters estimated from neutron-scattering experiments [6]. The interlayer coupling in Cs$_2$CuCl$_4$ appears to be smaller than $J$ and $J'$ by more than one order of magnitude, $J'' = 0.13$ K [9]. At $T_N = 0.62$ K, Cs$_2$CuCl$_4$ undergoes a phase transition into a cycloidal long-range-ordered state with an incommensurate wave vector $q = (0.0, 0.472, 0)$ [10]. Saturation fields of Cs$_2$CuCl$_4$ ($\mu_0 H_{sat} = 8.44, 8.89$, and 8 T, along the $a$, $b$, and $c$ axes, respectively [11]) can be reached using standard superconducting magnets, allowing one to experimentally investigate the phase diagram in detail [11,12]. The observation of a number of subsequent low-temperature field-induced transitions have triggered intensive theoretical studies ([5] and references herein), revealing the important role of $\mathcal{H}_\delta$ (Eq. 1). Very recently, several new field-induced transitions were ob-

![FIG. 1: Schematic view of exchange paths in Cs$_2$CuCl$_4$ with the exchange coupling $J$ along the $b$ axis (chain direction) and $J'$ along the zigzag bonds in the $bc$ plane.](image-url)
served in $\text{Cs}_2\text{CuCl}_4$, emerging under applied hydrostatic pressure $\text{[14]}$. Inelastic neutron-scattering experiments on $\text{Cs}_2\text{CuCl}_4$ revealed the presence of a highly dispersive continuum of excited states $\text{[12, 13]}$. These states have been initially identified as 2D RVB states, as suggested to occur in the AF system of spins on a triangular layer lattice $\text{[7]}$. However, later on, the data have been re-interpreted in the framework of the quasi-1D Tomonaga-Luttinger spin-liquid scenario $\text{[16]}$, with spinons (and their interchain bound-state excitations, triplons) as elementary magnetic excitations. A key condition here is the presence of the geometrical frustration, making spin chains well-isolated from each other (this is in contrast to the 2D Majorana spin-liquid scenario $\text{[17]}$, with magnetic excitations, however, coherently propagating along the direction of the strongest exchange interaction). The quasi-1D Tomonaga-Luttinger spin-liquid scenario perfectly describes the overall picture of magnetic excitations in $\text{Cs}_2\text{CuCl}_4$, including their behavior in magnetic fields $\text{[18]}$. ESR studies have supported the proposed model, with the uniform DM interaction opening an energy gap ($\sim 0.7$ K) at the $\Gamma$ point in the quantum-disordered state $\text{[19]}$. The thermal-conductivity experiments on $\text{Cs}_2\text{CuCl}_4$ were performed in a $^3\text{He}$-cryostat, in magnetic fields up to 14 T. Samples were prepared in a rod-like shape (with a thickness of about 0.5 mm and a length of about 5 mm) to allow for a sufficiently high temperature gradient. The temperature difference was produced by a heater attached at one end of the sample and measured by a pair of matched RuO$_2$ thermometers (as thermometers we used standard thick-film RuO$_2$-based SMD resistors); a four-point technique was employed to measure the temperature gradient along the sample. To reduce the statistical error, one point in the thermal conductivity (at fixed temperature and magnetic field) was averaged over 20 measurement cycles. In the experiments, the magnetic field was applied along the direction of the heat transport. The temperature in the entire field range was measured with accuracy better than $\pm 5\%$.

The thermal-conductivity experiments on $\text{Cs}_2\text{CuCl}_4$ revealed a broad maximum at about 5 K for all three directions (Fig. 2). Such a behavior is a text-book example of the phonon-dominated thermal conductivity, where the low-temperature conductivity is determined by phonon scattering on the crystal boundaries and structural imperfections, while the high-temperature mean-free path is limited by the Umklapp scattering $\text{[22]}$. As one can see, absolute values of the thermal conductivity (in particular at the maximum positions) are different for different directions: it is maximal for the direction along the $c$ axis and few times smaller for the $a$ and $b$ directions. This difference can be tentatively explained by the anisotropy of sound velocity and phonon-magnon scattering in $\text{Cs}_2\text{CuCl}_4$ at low temperatures.

With decreasing temperature we observed that the heat-transport behavior along the $b$ direction becomes significantly different from that along the $a$ and $c$ directions. Along the $b$ axis, before entering the 3D ordered phase, the thermal conductivity changes the slope (reflecting its pronounced enhancement) and suddenly drops, once the material undergoes the 3D ordering. The anomaly position depends on the applied magnetic field, shifting to lower temperatures with increasing field (which is consistent with the temperature-field phase diagram obtained by Tokiwa et al. $\text{[11]}$). Based on these observations, we suggest that the anomaly of the thermal conductivity, revealed by us in the vicinity of $T_N$ specifically along the $b$ direction, has a magnetic nature and is determined by magnetic excitations propagating along the direction of the largest exchange interaction (chain direction). For the Tomonaga-Luttinger
spin liquid, proposed for Cs$_2$CuCl$_4$ [16], these excitations are spinons. Similar anisotropic behavior of the thermal transport was observed in quasi-1D chain materials Sr$_2$CuO$_3$ [23, 24], SrCuO$_2$ [24], Cu(C$_4$H$_8$N$_2$)(NO$_3$)$_2$ [23], and CaCu$_2$O$_3$ [26]. Noticeably, apart from the shift, the applied magnetic field suppresses the anomaly, making it almost undetectable at 8 T. Such a behavior is consistent with the field-induced crossover from the quantum to a classically-favored state, where quantum fluctuations are significantly suppressed by magnetic field [27]. The sudden drop of the thermal conductivity $\kappa_b$ at $T_N$ can be explained by a collapse of the 1D spinon-heat transport when entering the 3D AF ordered state, accompanied by the opening of an energy gap in the low-temperature excitation spectrum. Such a gap ($\sim 1.3$ K) was observed in Cs$_2$CuCl$_4$ by means of ESR [28].

Field measurements revealed a non-monotonic behavior of the thermal conductivity for all three directions of the applied magnetic fields (Fig. 3). For the heat transport along the $b$ and $c$ axes, we observe changes in the heat transport behavior, which occur in magnetic fields about 2 T. These anomalies correspond to field-induced transitions from the 3D ordered to a disordered state, as reported previously [11, 13]. Noticeable low-field jumps of $\kappa_b$ at about 0.5 and 0.4 K (Fig. 3) provide additional evidences of the 1D nature of magnetic excitations in this material, significantly contributing to the heat transport just above $T_N$.

Another peculiar finding is the pronounced dip in the filed dependences of thermal conductivity at about 6-7 T, observed for all three directions (Fig. 3). This observation is consistent with the low-temperature ultrasound properties of Cs$_2$CuCl$_4$ [29–31], suggesting the exchange-striction mechanism as a possible reason of such a behavior. A pronounced increase of the thermal conductivity above $H_{sat}$ can be understood taking into account the decreased phonon-magnon scattering in the fully spin-polarized phase (the positions of corresponding maxima in thermal conductivity along the $b$ direction are shown in Fig. 4 by crosses). The phase diagram obtained by us, together with results of previously reported specific-heat and magnetic-susceptibility studies [11] for $H \parallel b$, is shown in Fig. 4; excellent agreement between the two data sets is found.

To estimate the magnetic mean-free path $l^m_b$ along the $b$ direction, low-temperature heat transport properties of Cs$_2$CuCl$_4$ were analyzed in the framework of the spin-1/2 AF Heisenberg chain model [26] with spinons as elementary magnetic excitations:

$$l^m_b = \frac{\kappa^m_b}{T} \frac{3\hbar}{N_s k_B^2 \pi}.$$  

Here, $\kappa^m_b$ is the magnetic heat conductivity, $\hbar$ is the reduced Planck constant, and $N_s = 4/ac$ is the number of spins per unit area. To estimate the magnetic heat conductivity $\kappa^m_b$ the nonmagnetic background $\kappa^{nm}_b$ was subtracted from the experimental data $\kappa_b$. To obtain $\kappa^{nm}_b$, the zero-field thermal-conductivity data $\kappa_b$ below and above the anomaly were fitted with the power law.
\[ \kappa = 0.1496 \cdot T^{1.875} \] (line \( \kappa_b^{nm} \) in Fig. 5). For comparison, the rescaled thermal conductivity along the \( a \) direction \( \kappa_a/2 \) (where no anomaly was observed) is shown, revealing excellent agreement with the fit results. The subtraction results yield 0.08 W/Km for the magnetic part of the heat conductivity at the maximum position. Based on this model, we obtained \( l_m \approx 200 \) Å, which corresponds to approximately 30 lattice spacings along the \( b \) direction.

In conclusion, the thermal conductivity in Cs\(_2\)CuCl\(_4\) was measured at temperatures down to 300 mK in magnetic fields up to 14 T along the three principal crystallographic directions. The heat transport is found to be dominated by the phonon contribution. On the other hand, a pronounced field-dependent anomaly of the thermal conductivity was observed along the \( b \) axis, when approaching the transition into the 3D ordered state. The anomaly is attributed to the 1D heat transport through magnetic excitations propagating in Cs\(_2\)CuCl\(_4\) along the direction of the strongest exchange coupling. Our observations strongly support the quasi-1D spin-liquid scenario with spinons as elementary excitations, proposed for this frustrated antiferromagnet.

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