Evidence of one-dimensional magnetic heat transport in the triangular-lattice antiferromagnet Cs_2CuCl_4

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We report on low-temperature heat-transport properties of the spin-1/2 triangular-lattice antiferromagnet Cs_2CuCl_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 found 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. In addition, peculiarities of the heat transport of Cs_2CuCl_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 [1–6]. 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 [7]. 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_2CuCl_4 has drawn a particularly great deal of attention. Magnetic Cu^{2+} ions in Cs_2CuCl_4 form a quasi-2D lattice with the exchange coupling parameters 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

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

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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 [8] 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 [9]. The interlayer coupling in Cs_2CuCl_4 is smaller than J and J' by more than one order of magnitude, $J'' = 0.13$ K [9]. At $T_N = 0.62$ K, Cs_2CuCl_4 undergoes a phase transition into a cycloidal long-range-ordered state with an incommensurate wave vector $q = (0, 0.472, 0)$ [10]. Saturation fields of Cs_2CuCl_4 ($\mu_0 H_{\text{sat}} = 8.44, 8.89$, 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 [11–13]. The observation of a number of subsequent low-temperature field-induced transitions has 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 observed in Cs_2CuCl_4 , emerging under applied hydrostatic pressure [14].

Inelastic neutron-scattering experiments on Cs_2CuCl_4 revealed the presence of a highly dispersive continuum of excited states [12,15]. These states were initially identified as 2D RVB states, as suggested to occur in the AF system of spins on a triangular layer lattice [7]. However, later on, the data were reinterpreted in the framework of the quasi-1D Tomonaga-Luttinger spin-liquid scenario [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

FIG. 1. Schematic view of exchange paths in Cs_2CuCl_4 with the exchange coupling J along the b axis (chain direction) and J' along the zigzag bonds in the bc plane.

well isolated from each other (this is in contrast to the 2D Majorana spin-liquid scenario [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 Cs_2CuCl_4 , including their behavior in magnetic fields [18]. ESR studies have supported the proposed model, with the uniform DM interaction opening an energy gap (~ 0.7 K) at the Γ point in the quantum-disordered state [19].

Here, we report on heat-transport studies of Cs_2CuCl_4 , with the main goal to probe the anisotropy of the thermal conductivity on the temperature scale $T \sim J/k_B$, where effects of the dimensionality of magnetic correlations should become relevant ([20] and references therein).

Single crystals of Cs_2CuCl_4 were grown by the slow evaporation of aqueous solution of CsCl and CuCl_2 in the mole ratio 2 : 1. The room-temperature crystallographic parameters ($a = 9.769$ Å, $b = 7.607$ Å, and $c = 12.381$ Å; space group $Pnma$) were confirmed by x-ray diffraction and are in good agreement with those reported previously [21].

Thermal-conductivity measurements were performed in a ^3He cryostat, in magnetic fields up to 14 T. Samples were prepared in a rodlike 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 surface mount device 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 Cs_2CuCl_4 revealed a broad maximum at about 5 K for all three directions (Fig. 2). Such a behavior is a textbook 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 [22]. As one can see, absolute values of the thermal conductivity (in particular at the maximum positions)

FIG. 2. Temperature dependence of the thermal conductivity κ_b (a), κ_a (b), and κ_c (c) along the b , a , and c axes, respectively, in selected magnetic fields. Lines are guides to the eye.

are different for different directions: the value is maximal for the direction along the c axis and a 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 Cs_2CuCl_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, in contrast to that for other directions) 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.* [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. For the Tomonaga-Luttinger spin liquid, proposed for Cs_2CuCl_4 [16], these excitations are spinons. Similar anisotropic behavior of the thermal transport was observed in quasi-1D chain materials Sr_2CuO_3 [23,24], SrCuO_2 [24], $\text{Cu}(\text{C}_4\text{H}_4\text{N}_2)(\text{NO}_3)_2$ [25], and CaCu_2O_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].

FIG. 3. Field dependence of the thermal conductivity κ_b (a), κ_a (b), and κ_c (c) at selected temperatures. The magnetic field was applied along the heat-transport directions. Lines are guides to the eye.

The sudden drop of the thermal conductivity κ_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 (~ 1.3 K) was observed in Cs_2CuCl_4 by means of ESR [28].

Field measurements revealed a nonmonotonic 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 κ_b at about 0.5 and 0.4 K [Fig. 3(a)] provide additional evidence 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 field dependencies 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_2CuCl_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.

FIG. 4. Phase diagram of Cs_2CuCl_4 for magnetic field applied along the b direction. Data from our thermal-transport measurements are shown by red solid circles, while specific-heat and susceptibility data from [11] are denoted by open circles. The crosses correspond to the thermal conductivity maxima in the fully spin-polarized phase above H_{sat} (Fig. 3). Lines are guides to the eye.

To estimate the magnetic mean-free path l_b^m along the b direction, low-temperature heat transport properties of Cs_2CuCl_4 were analyzed in the framework of the spin-1/2 AF Heisenberg chain model [26] with spinons as elementary magnetic excitations:

$$l_b^m = \left(\frac{\kappa_b^m}{T} \right) \left(\frac{3\hbar}{N_s k_B^2 \pi} \right). \quad (2)$$

FIG. 5. Temperature dependencies of the thermal conductivity κ_b along the b axis (rough experimental data; blue symbols), nonmagnetic background κ_b^{nm} as the result of the power-law fit (magenta line), magnetic contribution to the thermal conductivity κ_b^m as the result of the subtraction κ_b^{nm} from κ_b (red symbols), and rescaled thermal conductivity $\kappa_a/2$ along the a direction (green symbols).

Here, κ_b^m 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 κ_b^m the nonmagnetic background κ_b^{nm} was subtracted from the experimental data κ_b . To obtain κ_b^{nm} , the zero-field thermal conductivity data κ_b below and above the anomaly were fitted with the power law $\kappa = 0.1496T^{1.875}$ (line κ_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/K m 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_2CuCl_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_2CuCl_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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