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We present the ultra-low-temperature thermal conductivity measurements on single crystals of the prototypical charge-density-wave material $1T$-TaS_2, which was recently argued to be a candidate for quantum spin liquid. Our experiments show that the residual linear term of thermal conductivity at zero field is essentially zero, within the experimental accuracy. Furthermore, the thermal conductivity is found to be insensitive to the magnetic field up to 9 T. These results clearly demonstrate the absence of itinerant magnetic excitations with fermionic statistics in bulk $1T$-TaS_2 and, thus, put a strong constraint on the theories of the ground state of this material.

The quantum spin liquid (QSL), where strong quantum fluctuations obstruct long-range magnetic order even down to the absolute zero temperature, is one of the most elusive and exotic quantum state of matter [1–3]. In the QSLs, Mott physics plays a significant role in localizing electrons and forming $S = 1/2$ spins, as has been manifested in the study of high-temperature superconductors [4]. Experimentally, triangular-lattice organic compounds κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ [5–7] and EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ [8–10], together with kagome-lattice $ZnCu_3(OH)_6Cl_2$ [11–15] and $Cu_3Zn(OH)_6FBr$ [16], are typical examples of Mott-assisted QSL candidates. However, the features of the spin frustrations in the above systems are affected by other factors, such as structural deformations or the intersite mixture [3]. From this point of view, it is very meaningful to seek structurally perfect and enough clean systems for the realization of a QSL ground state.

$1T$-TaS_2 has been recently argued to be such an example [17, 18]. It is a layered material, and the only correlation-driven insulator among transition metal dichalcogenides [19]. As for the charge degree of freedom, $1T$-TaS_2 features a number of peculiar charge density wave (CDW) phases. Upon cooling, it turns into a metallic incommensurate CDW (ICCDW) phase below 550 K, a textured nearly commensurate CDW (NCCDW) phase below 350 K, and finally enters a commensurate CDW (CCDW) phase below 180 K [20]. The low-temperature CCDW phase is characterized by a $\sqrt{13} \times \sqrt{13}$ structure described as star-of-David clusters [20]. There is one unpaired electron per David-star due to energy gaps induced by the periodic lattice distortion [20]. At the same time, the electron correlation effects set in and localize this electron, leading to a Mott insulating state with $S = 1/2$ spins arranged on an ideal triangular lattice [21–23]. This is one of the few model spin configurations that may harbor the exotic QSL state, and exactly the one proposed by Anderson in his resonating-valence-bond model [24–26].

The possibility of the realization of a QSL in $1T$-TaS_2 has been proposed in Ref. [17]. By analyzing the existing data of this material, it was argued that $1T$-TaS_2 should be considered as a QSL, either a fully gapped Z_2 spin liquid or a Dirac spin liquid [17]. The muon spin relaxation (μSR) and nuclear quadrupole resonance (NQR) experiments have been performed on $1T$-TaS_2 single crystals [18]. No long-range magnetic order was detected from 210 K down to 70 mK by μSR. On the other hand, the NQR experiments reveal a gapless QSL-like behavior in part of the CCDW phase, from 200 K to $T_f = 55$ K. Below T_f, a novel quantum phase with amorphous tiling of frozen singlets emerges out of the QSL [18]. Meanwhile, another group performed polarized neutron diffraction and μSR measurements on $1T$-TaS_2 [27]. Their results indicate the presence of the short-ranged magnetic order below 50 K, and support the scenario that an orphan $S = 1/2$ spin moment is localized at the center of the David-star [27].

To find out what is the true ground state of bulk $1T$-TaS_2, it is essential to know the details of the low-lying elementary excitations. Ultra-low-temperature thermal conductivity measurement has proven to be a powerful technique in the study of low-lying excitations in QSL candidates [7, 9, 28]. Taking the spin-1/2 triangular-lattice Heisenberg antiferromagnets as example, the thermal conductivity result implied a possibility of a tiny gap opening in κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ [7], while highly mobile gapless excitations with fermionic statistics exist in EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ [9]. For the QSL candidate $YbMgGaO_4$, which has been studied extensively recently, no significant contribution of thermal conductivity from magnetic excitations was observed [28].

In this Rapid Communication, we report the ultra-low-temperature thermal conductivity measurement on a high-quality $1T$-TaS_2 single crystal down to 0.1 K. No significant contribution from magnetic excitations is detected at zero magnetic field. Furthermore, the thermal conductivity is found to be insensitive to magnetic fields up to 9 T. The absence of κ_0/T at all fields unambiguously demonstrates that no fermionic magnetic excitations with itinerant character exist in $1T$-TaS_2. We shall discuss the implications of our findings on the ground state of bulk $1T$-TaS_2.

The high-quality $1T$-TaS_2 single crystal was grown by the chemical vapor transport method [27]. The x-ray diffraction (XRD) measurement was performed on the $1T$-TaS_2 sample by using an x-ray diffractometer (D8 Advance, Bruker). The single crystal with a large natural surface was cut to a rectangular shape of $3.25 \times 0.72 \times 0.1$ mm^3. The large natural surface (3.25×0.72 mm^2) was determined to be the (001) plane by XRD, as shown in Fig. 1(a). A standard four probe method was used for both resistivity and thermal conductivity measurements. Contacts were made directly on this natural surface with silver paint. The resistivity was measured in a ^{4}He cryostat from 300 to 1.5 K. The thermal conductivity was measured in a dilution refrigerator, using a standard four-wire steady-state method with two RuO_2 chip thermometers, calibrated *in situ* against a reference RuO_2 thermometer. Magnetic fields were applied perpendicular to the large natural surface.

As shown in the inset of Fig. 1(a), $1T$-TaS_2 crystallizes in the CdI_2-type trigonal structure belonging to the P-$3m1$ space group [29]. It has a layered structure, in which each atomic layer is composed of one Ta layer sandwiched between two S layers in an octahedral arrangement [30]. Within the CCDW phase, 13 Ta atoms form a fully interlocked David-star cluster, where 12 peripheral Ta atoms shrink towards the central Ta atom. Such a deformation leads to the formation of a $\sqrt{13} \times \sqrt{13}$ triangular superlattice [20], as illustrated in the bottom left inset of Fig. 1(b). The temperature dependence of the resistivity $\rho(T)$ for the $1T$-TaS_2 single crystal measured on cooling and warming in zero magnetic field is plotted in Fig. 1(b). The hysteresis around 200 K indicates the occurrence of the first-order phase transition between the NCCDW phase and the CCDW phase. Below the transition temperature, the resistivity exhibits an insulating behavior. At low temperature, the increase of $\rho(T)$ with decreasing temperature is slower than an exponential rise, as seen in the top right inset of Fig. 1(b). All of these features are consistent with previous resistivity measurements on $1T$-TaS_2 [31, 32]. The inverse resistivity ratio $\rho(1.5\,\mathrm{K})/\rho(295\,\mathrm{K})$ is about 130, which is comparable with those measured previously [31, 32].

Figure 2(a) presents the in-plane thermal conductivity of the $1T$-TaS_2 single crystal at $H = 0$ T. In a solid, the contributions to thermal conductivity usually come from various quasiparticles, such as phonons, electrons, magnons, and spinons. For $1T$-TaS_2, the thermal conductivity from electrons (κ_e/T) at 1.5 K is estimated to be 6.13×10^{-5} mW K^{-2} cm^{-1} according to the Wiedemann-Franz law $\kappa_e/T = L_0/\rho(1.5\text{ K})$, with the Lorenz number $L_0 = 2.45 \times 10^{-8}$ W Ω K^{-2} and ρ(1.5 K) = 399.76 mΩ cm. The electron contribution becomes smaller upon further cooling and is negligible at ultra-low temperature, due to the insulating behavior of the resistivity. Therefore, the thermal conductivity at very low temperature can be fitted by $\kappa/T = a + bT^{\alpha-1}$, in which the two terms aT and bT^α represent the contributions from fermionic magnetic excitations (if they exist) and phonons, respectively [34, 35]. Because of the specular reflections of phonons at the sample surfaces, the power α in the second term is typically between 2 and 3 [34, 35]. The fitting of 0 T data below 0.35 K gives the residual linear term $\kappa_0/T \equiv a = 0.005 \pm 0.002$ mW K^{-2} cm^{-1} and $\alpha = 2.69$. Considering our experimental error bar $\pm$ 5 μW K^{-2} cm^{-1}, the κ_0/T of $1T$-TaS_2 at zero field

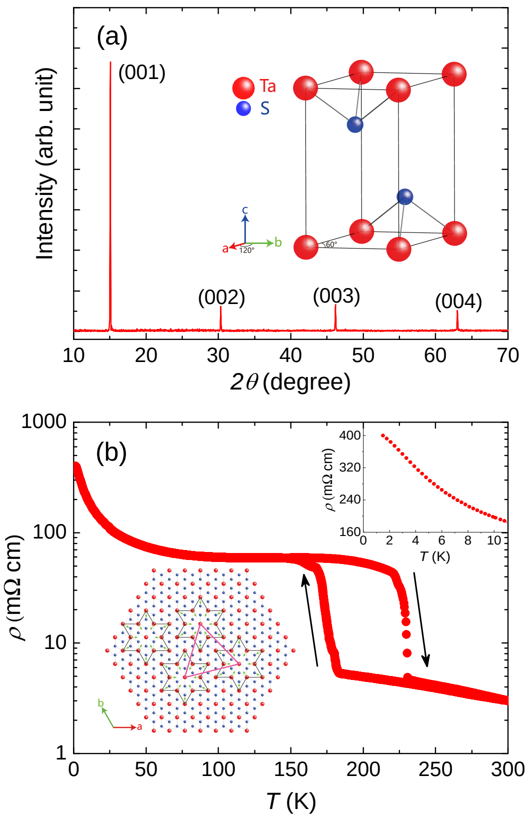

FIG. 1. (a) Room-temperature x-ray diffraction pattern from the large natural surface of the $1T$-TaS_2 single crystal. Only $(00l)$ Bragg peaks show up. Inset: trigonal P-$3m1$ crystal structure of $1T$-TaS_2. Ta (S) atoms are displayed by red (blue) balls. (b) Temperature dependence of the resistivity for the $1T$-TaS_2 single crystal measured on cooling and warming in zero magnetic field. The hysteresis can be clearly resolved around 200 K, suggesting the occurrence of the first-order phase transition between the NCCDW phase and the CCDW phase. Top right inset: the resistivity in the low-T region. Bottom left inset: a schematic of the monolayer $1T$-TaS_2 viewed along c axis in the CCDW phase. Black line indicates the representative David-star clusters, where in-plane Ta atom displacements are marked by green arrows. These David-star clusters further form the triangular superlattice (magenta line).

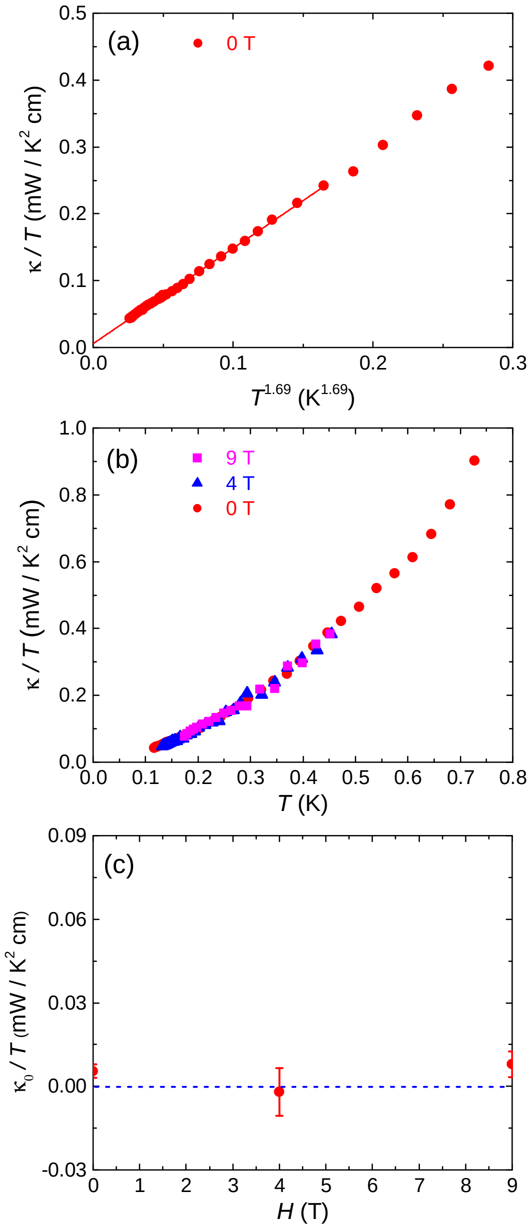

FIG. 2. (a) The in-plane thermal conductivity of the $1T$-TaS_2 single crystal at $H = 0$ T. The solid line represents the fit of the data to $\kappa/T = a + bT^{\alpha-1}$. This gives the residual linear term κ_0/T = 0.005 ± 0.002 mW K^{-2} cm^{-1} and $\alpha = 2.69$. (b) The in-plane thermal conductivity of $1T$-TaS_2 at various magnetic fields (H = 0, 4, and 9 T) applied along the c axis. (c) Field dependence of the residual linear term κ_0/T. The three κ_0/T values are negligible in our field range.

is essentially negligible. Note that $EtMe_3Sb[Pd(dmit)_2]_2$ has a value of κ_0/T as big as 2 mW K^{-2} cm^{-1} [9]. The in-plane thermal conductivity of the $1T$-TaS_2 single crystal in magnetic fields (H = 0, 4, and 9 T) applied along the c axis is plotted in Fig. 2(b), with the three curves almost overlapping on top of another. The same fitting process is performed, giving κ_0/T = -0.002 ± 0.009 mW K^{-2} cm^{-1} and κ_0/T = 0.008 ± 0.005 mW K^{-2} cm^{-1} for H = 4 and 9 T, respectively. The three κ_0/T values are plotted in Fig. 2(c). One can see that magnetic field barely has any effect on the thermal conductivity of $1T$-TaS_2 up to 9 T.

Now we would like to discuss the implications of our thermal conductivity results on the proposal of $1T$-TaS_2 being a QSL. Theoretically, all known QSLs can be classified in terms of a spectrum of gapless spinons (or their absence) and the nature of the emergent gauge fields to which they couple [36]. Various kinds of exotic models have been proposed in the study of various QSL candidates [3]. A systematic analysis of whether these models can be applied to $1T$-TaS_2 is beyond the scope of this work, and we only discuss the feasibility of these models in the light of our experimental data on the low-energy spin excitations. Generally, a finite residual linear term κ_0/T represents the contribution to κ from fermionic magnetic excitations in the zero temperature limit, i.e., the spectrum of the fermionic magnetic excitations is gapless. This might come from a spinon-Fermi surface or nodes in the momentum space. For $1T$-TaS_2, the former one has been ruled out [17], because of the tiny linear term γ ($\sim$ 2 mJ mol^{-1} K^{-2}) observed in specific heat [27, 37]. For the latter one, the most common case is a U(1) Dirac spin liquid [4, 38, 39]. In such a state, nodal fermionic spinons at the Dirac points would still result in a finite κ_0/T, and the thermal conductivity would be enhanced by a magnetic field [39]. This is incompatible with our results that the κ_0/T is negligible at all fields and the thermal conductivity is insensitive to magnetic field. It seems that any gapless QSL scenarios, whether gapless everywhere or only at nodes in the momentum space, are not consistent with our data. Note that there are also some exotic scenarios with nodal bosonic excitations [3, 40]. The contribution to the thermal conductivity from these nodal excitations exhibits a power-law temperature dependence ($\sim T^\delta$). However, unlike nodal fermionic excitations, for which the power-law exponent δ is 1, the δ value for nodal bosonic excitations is unknown in advance, so that it is hard to be separated from the phonon contribution.

However, there is another possibility that might reconcile our data with the gapless QSL scenarios. For the low-temperature phase ($T \leq T_f$ = 55 K) of $1T$-TaS_2, the NQR shows a broad distribution of $1/T_1$ values with a stretched exponent $p < 1$ ($p \approx 0.5$), implying a highly inhomogeneous magnetic phase at all Ta sites [18]. We note that similar spectral broadening and stretched exponent behavior have been observed in another triangular-lattice QSL candidate κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ [41]. The thermal conductivity measurement on κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ also gives a negligible κ_0/T, which was argued to come possibly from the localization of the gapless spin excitations due to the inhomogeneity [42]. This might also be the case for $1T$-TaS_2. The low-temperature phase ($T \leq T_f$) still exhibits a gapless behavior for the low energy fractional excitations, according to Ref. [18], but these gapless excitations can be localized so that they cannot conduct heat.

Next we turn to the gapped QSL scenarios. A fully gapped Z_2 spin liquid was suggested in Ref. [17], which is a state with gapped spinons together with gapped visons. For κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, an alternative explanation of the negligible κ_0/T is that the spin excitations are gapped [7]. The total thermal conductivity of κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ is the sum of a phonon contribution term and a magnetic part with an exponential temperature dependence, indicating the existence of a gap ($\Delta \sim 0.46$ K) in the spin excitation spectrum [7]. A field-induced gap closing was also observed in κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ for magnetic fields higher than ~ 4 T [42]. For 1T-TaS$_2$, however, we found that a similar fitting procedure does not work. Having said that, we caution that our data do not contradict the gapped QSL scenario. Indeed, if the magnitude of the gap is sufficiently large, the magnitude of the exponential term in the total thermal conductivity would be too small to be discerned at such low temperature, so that the total thermal conductivity is dominated by the phonon term. And the gap, if it exists, cannot be closed by a magnetic field up to 9 T. For example, an unusually large exchange interaction $J \approx 0.13$ eV (~ 1500 K) has been derived from the susceptibility data [18]. The spin gap is estimated to be above 200 K in Ref. [17]. In fact, in most cases a large gap is more common than a small gap (compared to its J) as possibly in κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ [7].

Of course, our results do not entirely rule out the possibility that the ground state of bulk 1T-TaS$_2$ may not be a QSL. The low-temperature phase ($T \leq T_f$) proposed by the NQR experiment is a highly unusual one, featuring frozen singlets, pseudogap in the spinon density of states, and a high degree of local disorder [18]. In Ref. [27], the neutron diffraction and μSR results provide evidence for the existence of a short-range-ordered state. To what extent do the behaviors of these states resemble those of a QSL is an open question and requires future scrutiny. As stated in Ref. [17], it might be more interesting to look for a QSL ground state in ultra-thin crystals of 1T-TaS$_2$.

In summary, we have measured the thermal conductivity of a 1T-TaS$_2$ single crystal down to 0.1 K. No residual linear term of thermal conductivity was observed at zero field. The thermal conductivity is found to be insensitive to a magnetic field up to 9 T. These results provide evidence for the absence of itinerant magnetic excitations obeying fermionic statistics in 1T-TaS$_2$. Our results set strong constraints on the nature of its ground state and, thus, of its theoretical description.

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