Editors' Suggestion

# Absence of Magnetic Thermal Conductivity in the Quantum Spin Liquid Candidate $EtMe_3Sb[Pd(dmit)_2]_2$

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We present the ultralow-temperature specific heat and thermal conductivity measurements on single crystals of triangular-lattice compound $EtMe_3Sb[Pd(dmit)_2]_2$, which has long been considered as a gapless quantum spin liquid candidate. In specific heat measurements, a finite linear term is observed, consistent with the previous work [S. Yamashita *et al.*, Nat. Commun. **2**, 275 (2011)]. However, we do not observe a finite residual linear term in the thermal conductivity measurements, and the thermal conductivity does not change in a magnetic field of 6 T. These results are in sharp contrast to previous thermal conductivity measurements on $EtMe_3Sb[Pd(dmit)_2]_2$ [M. Yamashita *et al.*, Science **328**, 1246 (2010)], in which a huge residual linear term was observed and attributed to highly mobile gapless excitations, likely the spinons of a quantum spin liquid. In this context, the true ground state of $EtMe_3Sb[Pd(dmit)_2]_2$ has to be reconsidered.

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Quantum spin liquid (QSL) states have been one of the central issues in condensed matter physics since the seminal proposal by Anderson [1,2]. Because of the strong geometrical frustration and quantum fluctuations, the spins do not order even down to zero temperature and remain highly entangled, and fractionalized excitations called spinons are the most pronounced characteristic in QSL states [3–5]. The detection and study of these excitations are of great importance to give information about the nature of QSL states. As the prototype of a QSL in Anderson's resonating valence bond model [1], the spin 1/2 triangular-lattice Heisenberg antiferromagnet is a platform for searching the QSL candidates. Among those triangular-lattice QSL candidates, the two organic compounds $\kappa$-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ and $EtMe_3Sb[Pd(dmit)_2]_2$ seem to be promising [6–16], while the inorganic compound $YbMgGaO_4$ is under hot debate [17–23].

$EtMe_3Sb[Pd(dmit)_2]_2$ (Me = $CH_3$, Et = $C_2H_5$, dmit = 1, 3-dithiole-2-thione-4,5-dithiolate) is a compound of the series of layered organic salts $\beta'$-$X[Pd(dmit)_2]_2$, where $X$ is a nonmagnetic cation, and $Pd(dmit)_2$ is highly dimerized forming the spin 1/2 anion $[Pd(dmit)_2]_2^-$ [24,25]. Each $Pd(dmit)_2$ layer is parallel to the $ab$ plane and is separated by the $EtMe_3Sb^+$ cation layer, as seen in Fig. 1(a) (cation layers are not shown for clarity). The dimerized $Pd(dmit)_2$ are arranged to form a triangular lattice in the layer, causing a strong geometrical frustration of spins on the dimers, illustrated in Fig. 1(b). No signature of long range magnetic order was observed down to about 20 mK by nuclear magnetic resonance (NMR) measurements [13,14], in spite of the large effective antiferromagnetic exchange interactions of the order of 250 K [11]. However, the deviation of NMR spin-lattice relaxation curves from single exponential functions below 10 K indicates that the spin state gradually becomes inhomogeneous [13,14]. As for the study of excitations in this putative QSL candidate, the spin-lattice relaxation $1/T_1$ was found to follow the $T^2$ temperature dependence below 1 K, indicating a nodal spin gap [13,14]. In contrast, a finite linear term $\gamma$ of 19.9 $mJ\,K^{-2}\,mol^{-1}$ was observed in the specific heat measurements, implying the existence of gapless fermionic excitations [15]. The gapless nature was further confirmed by the torque magnetometry, which revealed residual paramagnetic susceptibility comparable to that in metal [16].

More strikingly, the previous thermal conductivity work of $EtMe_3Sb[Pd(dmit)_2]_2$ single crystals by M. Yamashita *et al.* [12] reported the observation of a huge residual linear term $\kappa_0/T$ of 2 $mW\,K^{-2}\,cm^{-1}$, which is attributed to highly mobile gapless fermionic excitations with the mean free path as long as 1000 interspin distances. This is a strong evidence for the existence of a spinon Fermi surface in such a QSL candidate, stimulating a number of theoretical studies [4,5]. The enhancement of $\kappa$ above 2 T was also observed [12]. However, for the other two triangular-lattice QSL candidates $\kappa$-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ and $YbMgGaO_4$, no residual linear terms were found in thermal conductivity measurements [9,20], although they also display a gapless nature in specific heat measurements [i.e., a linear term $\gamma$ of 15 $mJ\,K^{-2}\,mol^{-1}$ in $\kappa$-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ and a power-law temperature dependence in $YbMgGaO_4$,

 

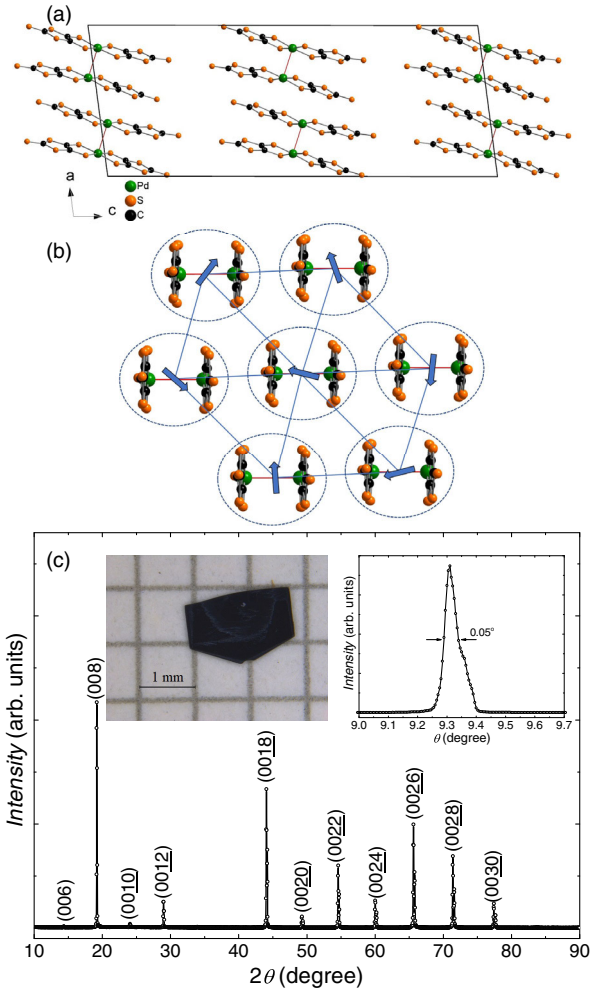


FIG. 1. (a) Crystal structure of $EtMe_3Sb[Pd(dmit)_2]_2$ with only the $Pd(dmit)_2$ layers shown. (b) The triangular-lattice structure of the spin 1/2 in the $Pd(dmit)_2$ layer. The dimerized $Pd(dmit)_2$ are surrounded by dashed ovals. Each 1/2 spin, denoted as a blue arrow, is localized on each dimer. (c) Room-temperature XRD pattern from the natural surface of the $EtMe_3Sb[Pd(dmit)_2]_2$ single crystal. Left inset: the photo of a typical $EtMe_3Sb[Pd(dmit)_2]_2$ single crystal. Right inset: x-ray rocking scan curve of the (008) Bragg peak.

respectively] [8,17,20]. One may raise the question why $EtMe_3Sb[Pd(dmit)_2]_2$ is so different from other triangular-lattice QSL candidates. Therefore, as the vital experimental foundation of many theoretical works, it is desired to revisit these thermodynamic and transport properties of $EtMe_3Sb[Pd(dmit)_2]_2$.

In this Letter, we report the ultralow-temperature specific heat and thermal conductivity measurements on high-quality $EtMe_3Sb[Pd(dmit)_2]_2$ single crystals. A linear term $\gamma$ of 15 mJ K$^{-2}$ cm$^{-1}$ is found in the specific heat, which is consistent with the previous work [15]. However, it is unsuccessful to reproduce previous thermal conductivity results reported in Ref. [12]. A negligible residual linear term $\kappa_0/T$ is observed, implying the absence of mobile gapless fermionic excitations. This raises the question about the true ground state of this triangular-lattice QSL candidate.

Single crystals of $EtMe_3Sb[Pd(dmit)_2]_2$ were provided by the same group at RIKEN as in Ref. [12]. The photo and the typical x-ray diffraction (XRD) pattern (measured by a D8 Advance x-ray diffractometer from Bruker) from the natural $ab$ plane of a $EtMe_3Sb[Pd(dmit)_2]_2$ sample are shown in Fig. 1(c). The full width at half-maximum (FWHM) of the x-ray rocking scan curve of the (008) Bragg peak is only 0.05°, indicating the high quality of the samples. The specific heat of a sample with $0.4 \pm 0.1$ mg was measured by the relaxation method in a physical property measurement system (Quantum Design) equipped with a $^3$He cryostat. Since stacking more samples together will lead to a bad thermal contact, we only measured the specific heat of one large sample with comparable size of the sample holder. The light mass of the sample is due to its low density and small thickness. Samples for thermal conductivity measurements have the dimensions of $1.34 \times 0.60 \times 0.05$ mm$^3$ for sample 1, $2.00 \times 1.17 \times 0.05$ mm$^3$ for sample 2, and $1.34 \times 0.88 \times 0.02$ mm$^3$ for sample 3, respectively. Note that the samples used for thermal conductivity measurements come from three different batches. All samples were prepared in almost the same conditions as samples in Ref. [12]. As for sample 3, especially, the conditions were the same including the reagents. Contacts of sample 1 were made using gold paste. Contacts of samples 2 and 3 were made using carbon paste. The thermal conductivity was measured in a dilution refrigerator, using a standard four-wire steady-state method. Samples were cooled slowly from room temperature to the lowest temperature for two days in order to avoid cracks. Magnetic fields were applied perpendicular to the $ab$ plane. A scanning electron microscope (SEM, Zeiss EVO-10) was used to examine the surface of sample 1 after thermal conductivity measurements.

The temperature dependence of the specific heat of $EtMe_3Sb[Pd(dmit)_2]_2$ single crystal from 0.65 to 4 K at zero field is shown in the inset of Fig. 2. The main panel of Fig. 2 plots the $C/T$ vs $T^2$ below 2 K. The upturn at low temperatures is attributed to the Schottky anomaly [15]. $C/T$ can be well fitted by the formula $C/T = \gamma + \beta T^2$ between 0.9 and 2 K, giving $\gamma = 15 \pm 4$ mJ K$^{-2}$ mol$^{-1}$ and $\beta = 17 \pm 4$ mJ K$^{-4}$ mol$^{-1}$ after considering the error in mass. This behavior is similar to that in Ref. [15], which is also plotted in Fig. 2. Actually, if we scale our data by a factor of 1.36, they are on top of the curve in Ref. [15]. Thus, the previous specific heat result is well reproduced by us, and the slight difference is likely due to the uncertainty in determining the mass of samples.

Since a specific heat result may be contaminated by nuclear contributions [8,15], the thermal transport measurement, a tool insensitive to localized excitations, would be more advantageous for identifying the

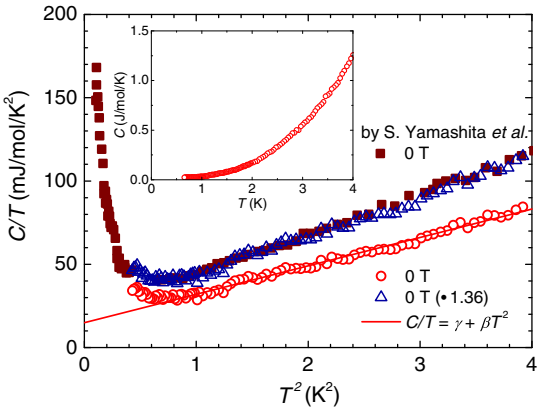


FIG. 2. Temperature dependence of the specific heat of $EtMe_3Sb[Pd(dmit)_2]_2$ single crystal at zero field. The inset shows $C$ vs $T$ from 0.65 to 4 K. The main panel plots $C/T$ vs $T^2$ below 2 K. The solid line is the fit to the specific heat data between 0.9 K and 2 K to $C/T = \gamma + \beta T^2$. A linear term $\gamma = 15 \pm 4\ \mathrm{mJ\,K^{-2}\,mol^{-1}}$ is obtained. For comparison, the specific heat result in Ref. [15] is also plotted. Scaling by a factor of 1.36 can well reproduce the prior result, indicating that the difference is likely due to the uncertainty in measuring the mass of the samples.

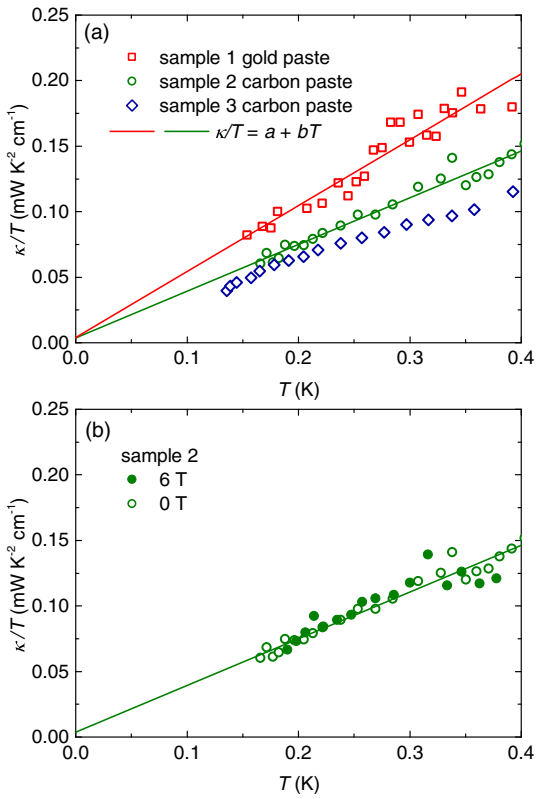


FIG. 3. (a) The zero-field in-plane thermal conductivity of three $EtMe_3Sb[Pd(dmit)_2]_2$ single crystals, using contacts of gold paste and carbon paste, respectively. (b) The in-plane thermal conductivity of sample 2 at $\mu_0 H = 0$ and 6 T. The straight lines are the fits to the zero-field thermal conductivity data to $\kappa/T = a + bT$.

low-energy excitations in a QSL candidate. Figure 3(a) presents the in-plane zero-field thermal conductivity of $EtMe_3Sb[Pd(dmit)_2]_2$ single crystals with two kinds of contacts, respectively. The thermal conductivity of an insulating QSL candidate at very low temperatures usually can be fitted by $\kappa/T = a + bT^{\alpha-1}$, in which the two terms $aT$ and $bT^{\alpha}$ represent the contributions from itinerant gapless fermionic magnetic excitations (if they do exist) and phonons, respectively. For phonons, the power $\alpha$ is typically between two and three, due to the specular reflections at the sample surfaces [26,27]. As can be seen in Fig. 3(a), similar thermal conductivity results between samples with gold and carbon pasted contacts are observed. Furthermore, contrary to Ref. [12], where $\kappa/T$ shows $T^2$ temperature dependence, a linear fitting $\kappa/T = a + bT$ is more suitable for our samples 1 and 2. The fitting gives the value of $\kappa_0/T$ of $0.004 \pm 0.009\ \mathrm{mW\,K^{-2}\,cm^{-1}}$ and $0.004 \pm 0.005\ \mathrm{mW\,K^{-2}\,cm^{-1}}$ for samples 1 and 2, respectively. Considering the experimental error bar of $5\ \mu\mathrm{W\,K^{-2}\,cm^{-1}}$, the $\kappa_0/T$ of both samples at $\mu_0 H = 0$ T are virtually zero. The behavior of sample 3 is very similar to samples 1 and 2. Because of the slightly sublinear temperature dependence, it extrapolates to a small negative value of $\kappa_0/T$. Therefore, previous huge $\kappa_0/T$ of $EtMe_3Sb[Pd(dmit)_2]_2$ in Ref. [12] can not be reproduced in any of our samples.

The in-plane thermal conductivities of sample 2 at $\mu_0 H = 0$ and 6 T are plotted in Fig. 3(b). The magnetic field barely has any effect on the thermal conductivity of $EtMe_3Sb[Pd(dmit)_2]_2$ at 6 T. This magnetic field dependence, again, is in stark contrast to the previous measurements that report a gradual increase of thermal conductivity above approximately 2 T below 1 K [12]. For example, the observed magnetothermal conductivity $[\kappa(H) - \kappa(0T)]/\kappa(0\ \mathrm{T})$ is larger than 20% at 6 T at 0.23 K, which was suggested as a consequence of additional excitations with a spin gap closing at 2 T [12]. Because of the coexistence of gapless and gapped excitations, a type II spin liquid was proposed for $EtMe_3Sb[Pd(dmit)_2]_2$ [28]. Considering the negligible field effect presented here, one may reexamine the existence of this gapped excitation and the proposal of type II spin liquid.

Figure 4 displays the comparison of our thermal conductivity data with the data of $EtMe_3Sb[Pd(dmit)_2]_2$ and $Et_2Me_2Sb[Pd(dmit)_2]_2$ in Ref. [12] and $\kappa$-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ in Ref. [9]. The absolute value of our data are much smaller than that in Ref. [12], even 10 times smaller than the nonmagnetic reference compound $Et_2Me_2Sb[Pd(dmit)_2]_2$ [12]. The lack of $\kappa_0/T$, a negligible field effect in $\mu_0 H = 6$ T, and very small absolute value, all demonstrate that the thermal conductivity is entirely contributed by phonons in our samples, and the phonons are strongly scattered. This is supported by the estimation of the mean free path $l_p$ of phonons by the kinetic formula

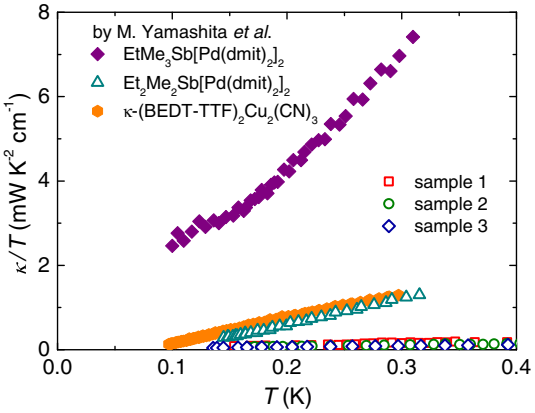


FIG. 4. Comparison of our thermal conductivity data with the data of $EtMe_3Sb[Pd(dmit)_2]_2$ and $Et_2Me_2Sb[Pd(dmit)_2]_2$ in Ref. [12] and $\kappa$-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ in Ref. [9]. The absolute value of our data are much smaller than that in Ref. [12], even 10 times smaller than the nonmagnetic reference compound $Et_2Me_2Sb[Pd(dmit)_2]_2$, indicating phonons being strongly scattered in our samples.

$\kappa = \frac{1}{3} C_p v_p l_p$, where $C_p = (2\pi^2 k_B/5)(k_B T/\hbar v_p)^3 = \beta T^3$ is the phonon specific heat and $v_p$ is the velocity of phonons. With $\beta = 17\ \mathrm{mJ\,K^{-4}\,mol^{-1}}$ obtained by our specific heat measurement, $v_p$ is estimated as $1.55 \times 10^3$ m/s, giving $l_p$ of sample 3 only about 5.88 $\mu$m at 0.3 K. Usually, when the sample enters the boundary scattering limit at sub-Kelvin temperatures, $l_p$ should only be limited by the physical dimensions of the sample, resulting in a temperature independent $l_p = 2\sqrt{A/\pi}$, where $A$ is the cross-sectional area of the sample [27]. For sample 3, the boundary limited $l_p$ is 150 $\mu$m. This is 25 times larger than $l_p$ estimated from our thermal conductivity data and is consistent with the fact that the thermal conductivity of magnetic $EtMe_3Sb[Pd(dmit)_2]_2$ is 10 times lower than the nonmagnetic $Et_2Me_2Sb[Pd(dmit)_2]_2$. It is likely that the phonons are strongly scattered by those frustrated spins in $EtMe_3Sb[Pd(dmit)_2]_2$. Note that, in another triangular-lattice system, the thermal conductivity of magnetic $YbMgGaO_4$ is about half of the nonmagnetic $LuMgGaO_4$, again showing the scattering of phonons by the spins [20].

Now, we would like to discuss the implications of the negligible $\kappa_0/T$ in $EtMe_3Sb[Pd(dmit)_2]_2$ according to our new results. A gapless QSL may have fermionic spinons, which can form a Fermi surface like a metal, and provide a large density of states at low energies. Therefore, a sizable $\kappa_0/T$ is expected [29–32]. In Ref. [12], from the observed huge $\kappa_0/T = 2\ \mathrm{mW\,K^{-2}\,cm^{-1}}$, the authors estimated the mean free path $l_s$ of gapless fermionic excitations as long as 1000 interspin distances. However, according to the fitting result $\kappa_0/T = 0.004\ \mathrm{mW\,K^{-2}\,cm^{-1}}$ of our samples 1 and 2, an upper bound of $l_s$ around 10 Å is estimated, comparable to the interspin distance. In this context, the negligible $\kappa_0/T$ observed in our experiments is inconsistent with the existence of highly mobile gapless fermionic excitations and spinon Fermi surface in $EtMe_3Sb[Pd(dmit)_2]_2$. The reason for such a huge discrepancy between our results and those in Ref. [12] is not clear for us, but it is clearly not due to intrinsic sample dependence since we have achieved similar results in three samples from different batches, especially including one prepared from the same conditions including the reagents as those in Ref. [12]. Also, it is not caused by the extrinsic factor of microcracks, since we do not detect microcracks on sample 1 with SEM after thermal conductivity measurements. Below, we only discuss the feasibility of possible scenarios in light of our experimental data on low-lying excitations.

One possible scenario is that $EtMe_3Sb[Pd(dmit)_2]_2$ has a fully gapped ground state. This scenario was initially used to explain the absence of $\kappa_0/T$ in $\kappa$-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ [9]. By fitting the sublinear temperature-dependent $\kappa/T$ at low temperature, a gap of about 0.46 K was obtained [9]. However, such a gapped scenario is inconsistent with the finite linear term in the specific heat. Therefore, the authors in Ref. [9] pointed out that the heat capacity measurements incorrectly suggest the presence of gapless excitation, possibly owing to the large Schottky contribution at low temperatures. Now that similar phenomena appear in $EtMe_3Sb[Pd(dmit)_2]_2$, one may take both compounds into account for this scenario.

Another possible scenario is that gapless excitations do exist in $EtMe_3Sb[Pd(dmit)_2]_2$ and $\kappa$-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$, indicated by specific heat measurements, but they are localized. Organic molecular compounds are generally thought of as extremely clean with a very low level of crystallographic defects. However, the exponent in the stretched-exponential fitting of the relaxation curve in NMR measurements of $\kappa$-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ deviates from unity substantially below 6 K, indicating some inhomogeneity therein [7]. This scenario was considered as an alternative explanation for the negligible $\kappa_0/T$ in $\kappa$-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ [33]. For $EtMe_3Sb[Pd(dmit)_2]_2$, the inhomogeneity was observed below 10 K in NMR measurements [13,14]. Although the stretching exponents which characterize the degree of inhomogeneity start to recover towards the homogeneous value below 1 K, they are only about 0.6 which still deviates from the value of a homogeneous system [13,14]. Thus, the inhomogeneity still cannot be ruled out in the sub-Kelvin temperature region [5]. Therefore, our thermal conductivity results raise the question regarding to what extent the inhomogeneity plays a role in the heat transport of $EtMe_3Sb[Pd(dmit)_2]_2$.

Finally, a proposed random-singlet state, based on the quenched disorder on spin-1/2 quantum magnets, can induce the gapless QSL-like state [34–36]. The linear temperature dependence of specific heat may be an evidence for a power-law density of states of randomness [35,36]. Interestingly, the linear or sublinear temperature

dependence of our thermal conductivity $\kappa/T$ is in agreement with the theoretical prediction for a random-singlet state [35]. This power law is a consequence of the scattering of acoustic phonons by quantum two-level systems from the distribution of random singlets [35], which may account for the huge suppression of our thermal conductivity shown in Fig. 4. However, we also notice that the specific heat is insensitive to the magnetic field and the susceptibility goes to a constant at low temperatures for $EtMe_3Sb[Pd(dmit)_2]_2$ [11,15], which are not consistent with the random singlet model.

In summary, we have revisited the thermodynamic and heat transport properties of triangular-lattice organic QSL candidate $EtMe_3Sb[Pd(dmit)_2]_2$. A linear term in the specific heat is well reproduced, as in the previous report [15], but the thermal conductivity shows a completely different behavior from Ref. [12]. No residual linear term $\kappa_0/T$ is observed at the zero-temperature limit, suggesting the absence of mobile gapless fermionic excitations in $EtMe_3Sb[Pd(dmit)_2]_2$. A magnetic field of 6 T does not affect the thermal conductivity, and its absolute value is even 10 times smaller than the nonmagnetic reference compound $Et_2Me_2Sb[Pd(dmit)_2]_2$. We conclude that there is no magnetic thermal conductivity but only the phonon thermal conductivity in $EtMe_3Sb[Pd(dmit)_2]_2$, and the phonons are strongly scattered by the frustrated spins. The absence of reproducible $\kappa_0/T$ in any QSL candidates so far presents a direct challenge to the realization of a gapless QSL with highly mobile spinons in frustrated quantum magnets.

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*Note added.*—We are aware of a similar thermal conductivity study of $EtMe_3Sb[Pd(dmit)_2]_2$ single crystals by the Taillefer group in Sherbrooke [37].

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