Systematic study on thermal conductivity of organic triangular lattice systems β'-X[Pd(dmit)$_2$]$_2$

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Systematic variation of the low-temperature heat transport properties of the quasi-two-dimensional quantum spin liquid (QSL) candidate β'-EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ and its analog compounds showing antiferromagnetic (AFM) ordering is discussed using thermal conductivity κ measurements. All compounds with the QSL ground state show a monotonic decrease in κ with decreasing temperature, which is a typical feature of glass or amorphous materials, caused by their low-energy phonon structures. In contrast, the AFM compounds exhibit a peak structure of κ around 10 K, which is the characteristic temperature dependence of normal crystals with coherent phonons. Our results indicate that the anomalous glassy behavior in β'-EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ is an intrinsic and universal feature of the organic QSL phase and is not caused by extrinsic factors, such as the quality of the samples and the microcracks occurring during the cooling process. We propose a scenario for the origin of the glassy behavior of QSL compounds in which the charge fluctuation within the Pd(dmit)$_2$ dimers changes the phonon properties from crystalline to glasslike.

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I. INTRODUCTION

A quantum spin liquid (QSL) emerging in Mott-Hubbard systems is an intriguing topic in condensed-matter physics [1]. It generally appears in localized spin-1/2 systems, with relatively large antiferromagnetic (AFM) interactions with $J = 10^{2-3}$ K, in the specific situation where geometrical frustration or the competition of different interactions with similar magnitudes prevents a long-range ordered structure. The spin degrees of freedom in a highly degenerate quantum mechanical ground state are left with fluidity and retain itinerancy near $T = 0$ K.

Organic charge-transfer complexes with a triangular lattice and inorganic kagome lattice systems have been considered promising candidates for QSL formers, and their magnetic ground state and elemental excitation have been studied extensively both theoretically and experimentally [2]. The organic Mott insulator β'-EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ (hereafter referred to as EtMe$_3$Sb salt) is a notable example of an organic QSL candidate with spin $S = 1/2$ [3,4], where Me = CH$_3$, Et = C$_2$H$_5$, and dmit = 1,3-dithiole-2-thione-4,5-dithiolate. Nuclear magnetic resonance (NMR) studies of this compound revealed the absence of magnetic ordering down to approximately 20 mK, even though the value of J is estimated to be $J/k_B \sim -250$ K by magnetic susceptibility measurements, which is experimental evidence of the QSL state in the EtMe$_3$Sb salt [5]. The heat capacity measurement demonstrated the presence of a residual liner term with the Sommerfeld constant $\gamma \sim 20$ mJ K^{-2} mol^{-1}, similar to Fermi liquid metals. This indicates a gapless excitation called a spinon in this compound [6]. Such fermionic residual terms in the heat capacity are widely observed even in other organic and inorganic QSL candidates, such as κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, κ-H$_3$(Cat-EDT-TTF)$_2$, ZnCu$_3$(OH)$_6$Cl$_2$, and YbMgGaO$_4$ [7–10]. The presence of a finite γ, even in the insulating and charge-gapped ground states, indicates the characteristic spin excitation of the EtMe$_3$Sb salt. However, the dynamical feature of the interacting spins in the QSL state of the EtMe$_3$Sb salt is still under debate.

Thermal conductivity κ measurement is a powerful tool for investigating the nature of the excitation in QSL formers [11], especially for determining whether the excitation is mobile or localized. In a previous study on κ of the EtMe$_3$Sb salt at extremely low temperatures below 300 mK, the significantly large residual term $\kappa_0/T \sim 0.2$ W K^{-2} m^{-1} as $T \to 0$ was observed when plotted as κ/T versus T^2, while the nonmagnetic analog compound Et$_2$Me$_2$Sb[Pd(dmit)$_2$]$_2$ showed no residual term [12]. This result suggests the mobile character of the spinon in the EtMe$_3$Sb salt. However, other groups recently reported a much smaller κ_0/T in the EtMe$_3$Sb salt, suggesting that the dominant contribution of κ is the phonon contribution κ_{ph} at low temperatures and the spin contribution is negligibly small [13,14]. It has been argued that the origin of this mismatch is the discrepancy of long-range coherence in the electronic state caused by the disorder of molecules or cracks in the crystals occurring during the cooling process [15]. The cooling rate dependence of κ was investigated; however, both its presence and absence have been claimed in recent reports [13,16]. There may be consensus in recent papers that κ of the EtMe$_3$Sb salt shows a glassy temperature dependence $\kappa \propto T^2$ at low temperatures, but the origin is still an open question. If this glassy character is induced by extrinsic factors such as a microcrack,

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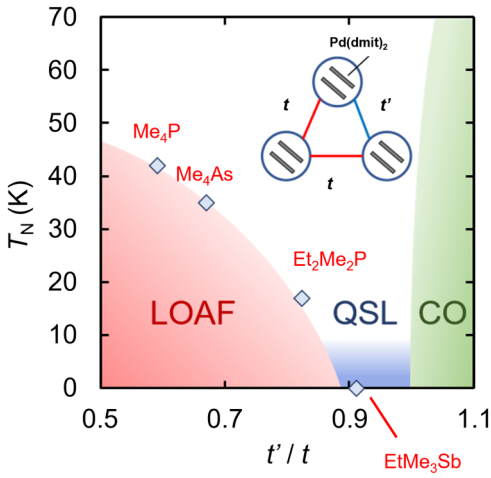

FIG. 1. Phase diagram of electronic and magnetic states of the β'-X[Pd(dmit)$_2$]$_2$ series as a function of the frustration parameter t'/t. The QSL state appears between the long-range antiferromagnetic order (LOAF) phase and the nonmagnetic charge order (CO) phase.

which is not related to the magnetic ground state, spin thermal conduction may be observed in disorder-free samples. However, if the QSL state is intrinsically associated with the glassy character of thermal conduction, it might be difficult to observe spin thermal conduction regardless of the quality of the sample and the measurement conditions such as the cooling rate because the mean free path of the spinon must be strongly suppressed. Therefore, whether the glassy behavior of κ of the EtMe$_3$Sb salt is intrinsic or extrinsic is important for understanding the nature of organic QSL formers more accurately.

A comparative experiment with an analog compound with different magnetic ground states is an effective way to clarify whether κ of the EtMe$_3$Sb salt is an intrinsic feature of QSL formers. As reported by Kato and Hengbo, the magnetic properties of β'-X[Pd(dmit)$_2$]$_2$ can be systematically tuned from an AFM to a QSL state by changing the counter cation X, namely, by tuning the hopping parameter t'/t, as shown in Fig. 1 [17]. Thus, it is possible to investigate the effect of the magnetic ground state on the temperature dependence of κ in the β'-X[Pd(dmit)$_2$]$_2$ series.

In this paper, we report a systematic study of κ of the β'-X[Pd(dmit)$_2$]$_2$ series in order to unveil the peculiar heat transport features inherent in the QSL ground state. Our results demonstrate the strong relationship between the magnetic state and thermal transport properties of phonons and the universal glassy feature of the organic QSL candidates.

II. EXPERIMENTAL DETAILS

Five different compounds of β'-X[Pd(dmit)$_2$]$_2$ with different counter cations X were selected. X = Me$_4$P, Me$_4$As, and Et$_2$Me$_2$P salts show AFM ordering at T_N = 42, 35, and 17 K, respectively [18]. In contrast, the EtMe$_3$Sb salt and Et(CD$_3$)$_3$Sb[Pd(dmit)$_2$]$_2$ [Et(CD$_3$)$_3$Sb salt] show QSL states even at extremely low temperatures. All the single crystals we measured were grown at RIKEN by the air oxidation method [17]. Thermal conductivity measurements using a single crystal were performed using the standard four-terminal steady flow method. The steady heat flow Q was created in the two-dimensional plane of the crystals (ab plane). The temperature gradient ΔT of the measurement was set to ~5% of the measurement temperature. We attached the different surface positions of single crystals to the heat sink (copper block), heaters (10 kΩ RuO$_2$ chips), and thermometers (10 kΩ RuO$_2$ chips or Cernox-1030 thermometers) by using gold wires (ϕ = 20 μm) and gold paste to detect ΔT in the samples. The cooling rate τ was ~0.1 K min^{-1} for all samples. In addition to the EtMe$_3$Sb data measured under these conditions (sample A), we also compared the data obtained at a different cooling rate of ~0.05 K min^{-1} by using a different EtMe$_3$Sb sample (sample B) from the same batch with sample A. After the measurements, we confirmed the absence of cracking on the surfaces of the samples. Thermal conductivity was calculated by the equation $\kappa = (QL)/(\Delta TA)$, where L and A are the length and cross-sectional area of the samples. The dimensions of the samples are shown in Table S1 in the Supplemental Material [19].

III. RESULTS AND DISCUSSION

The temperature dependences of the thermal conductivity κ of the antiferromagnet Me$_4$P, Me$_4$As, the Et$_2$Me$_2$P salt, and the QSL candidates EtMe$_3$Sb and Et(CD$_3$)$_3$Sb salt are shown in Fig. 2(a). The absolute values of κ for these compounds at 20 K show a discrepancy ranging between 3.3 and 0.1 W K^{-1} m^{-1} depending on their electronic ground state. These values are on the order of those previously reported for other organic conductors. The κ values of α-(BEDT-TTF)$_2$I$_3$ and several θ-(BEDT-TTF)$_2X$ [X = RbZn(SCN)$_4$, CsZn(SCN)$_4$] are 0.4–0.7 W K^{-1} m^{-1} at 20 K, resulting in a similar order of thermal conductivity [20,21]. Even for metallic compounds with itinerant electrons, the values range from 1 to 3 W K^{-1} m^{-1} around 20 K [22–25]. Because the electronic ground state of the measured compounds is a Mott insulating state at low temperatures, the electron/hole carrier contribution κ_{el} can be ignored in all samples. In the case of AFM salts, the spin contribution κ_{spin} is considered to be negligibly small at least below T_N. The absolute values of κ of the QSL candidates are significantly smaller than those of the AFM salts in all temperature ranges, suggesting the absence of a large κ_{spin}, at least in the high-temperature region. Therefore, the phonon contribution κ_{ph} is the dominant carrier of heat in all the samples.

As shown in Fig. 2(a), the temperature dependence of the thermal conductivity of Me$_4$P, Me$_4$As, and Et$_2$Me$_2$P increases with decreasing temperature at more than 10 K and then decreases monotonically, showing a broadened peak structure around 5–15 K. In general, κ_{ph} is described by the formula $\kappa_{ph} = 1/3 C_{ph} \nu_{ph} l_{ph}$, where $C_{ph} = \beta T^3$ is the lattice heat capacity (Debye's law), $\nu_{ph} = k_B/\hbar\sqrt[3]{2\pi^2 k_B/5\beta}$ is the sound velocity, and l_p is the mean free path of phonons. For a normal crystal, the temperature dependence of κ_{ph} in the high-temperature region is dominated by the temperature dependence of l_{ph}, which usually follows T^{-1} dependence (Euken's law). In contrast, κ_{ph} in the low-temperature region is mainly determined by the temperature dependence of C_{ph}.

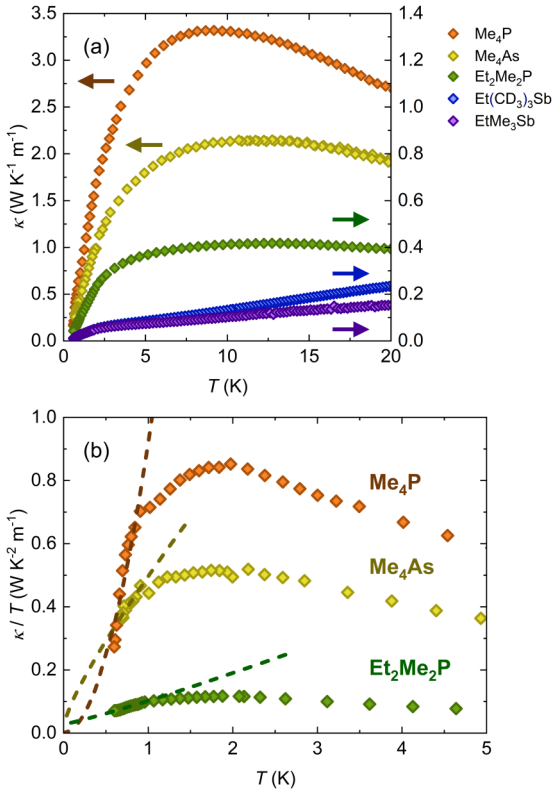

FIG. 2. (a) Temperature dependence of the thermal conductivity κ of the β'-X[Pd(dmit)$_2$]$_2$ series shown as a κ vs T plot. (b) κ/T vs T plot of the antiferromagnetic salts below 5 K. The dashed lines denote the free power-law fitting curve $\kappa/T = a + bT^c$ for the data below 1 K. The fitting parameter is shown in Table S1.

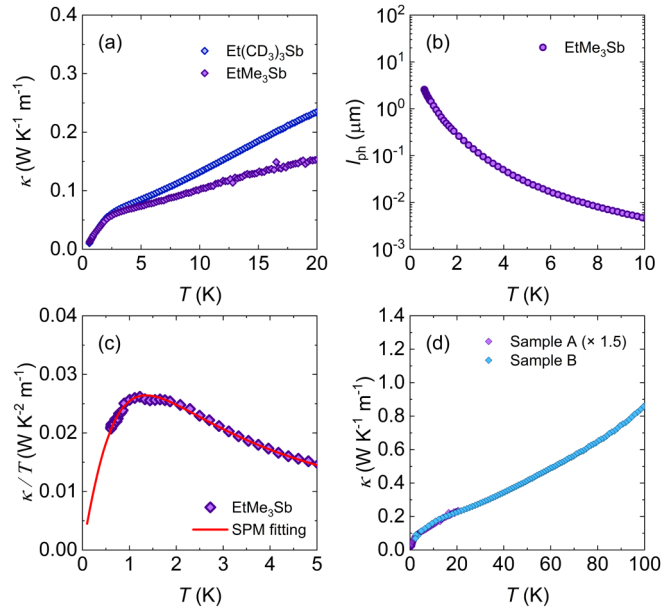

FIG. 3. (a) Temperature dependence of κ of the EtMe$_3$Sb salt (sample A) and the Et(CD$_3$)$_3$Sb salt shown as a κ vs T plot. (b) Temperature dependence of the mean free path of phonons of sample A. (c) κ/T vs T plot of sample A below 5 K. The red solid lines are the fitting curves of the soft-potential model where the fitting parameter $A = 0.0285$. (d) Temperature dependence of κ of two EtMe$_3$Sb salts, samples A and B, between 2 and 100 K. The absolute value of sample A is multiplied by 1.5 in this plot.

The temperature dependence of κ for the Me$_4$P salt almost follows the standard theory of κ. The Me$_4$As and Et$_2$Me$_2$P salts also show a similar trend in that they exhibit a peak structure of κ around 5 – 15 K, although their κ values are suppressed compared with that of the Me$_4$P salt. Comparing the temperature dependence of κ among these AFM compounds, the peak structure of κ is sharper, and the absolute value of the peak is on the order of $\kappa_{\mathrm{Me_4P}} > \kappa_{\mathrm{Me_4As}} > \kappa_{\mathrm{Et_2Me_2P}}$. Because the magnitude of the peak is related to l_{ph}, our results suggest that the mean free path of phonons in the AFM region is suppressed with an increasing of t'/t corresponding to the strength of spin frustration.

One possible reason for the suppression of phonon thermal conduction is the strong spin-phonon interaction. There have been several reports on low-dimensional intermetallic compounds in which spin-phonon scattering affects the temperature dependence of κ. For example, κ of $Tb_2Ti_2O_7$, which shows no magnetic ordering down to ultralow temperatures, is strongly suppressed compared with that of the nonmagnetic analog $Y_2Ti_2O_7$ [26]. Such spin-phonon scattering has been discussed for several materials such as $YbMgGaO_4$, α-$RuCl_3$, and $SrCu_2(BO_3)_2$ [10,27,28]. If the main phonon scattering factor is caused by the spin-phonon interaction, κ_{ph} is expected to change at the AFM transition temperature [29–33]. In the present case, however, no clear anomaly was observed in κ of the Et$_2$Me$_2$P salt at $T_{\mathrm{N}} = 17$ K. Thus, it is speculated that the effect of the spin-phonon interaction on the temperature dependence is small. In addition, the impurity (orphan) spins spontaneously emerging in the QSL state possibly induce such glassy properties [34,35]. However, a systematic temperature dependence of the thermal conductivity was observed even in the AFM long-range ordered state. Therefore, the disordering feature of the thermal properties cannot be attributed to the spin origin.

Figure 2(b) shows a κ/T versus T plot of these samples in the low-temperature region. A large peak with a maximum of approximately 0.8 W K^{-2} m^{-1} can be seen around 2 K but decreases to zero. Based on the free power-law fitting $\kappa/T = a + bT^c$ below 1 K (see the Supplemental Material [19]), the finite value κ_0/T can be estimated as 0–0.02 W K^{-2} m^{-1}. Such small values are reasonable for AFM ordered compounds since the magnetic ground state has a spin gap.

In contrast to the AFM compounds, the temperature dependences of κ of the EtMe$_3$Sb salt (sample A) and the Et(CD$_3$)$_3$Sb salt shown in Fig. 3(a) seem to show smaller κ values. For these QSL compounds, κ decreases with decreasing temperature, which is almost proportional to T, and this monotonic decrease becomes steeper at temperatures of less than 2 K. These temperature dependences of κ are clearly different from those of the AFM compounds and similar to those of glass or amorphous materials such as silica glass and organic polymers [36]. Here, the sound velocity of the EtMe$_3$Sb salt is estimated to 1370 m s^{-1}, calculated from the literature value $\beta = 24.1$ mJ K^{-4} mol^{-1} [6]. Thus, the temperature dependence of the mean free path of phonons l_{ph} of the EtMe$_3$Sb salt can be estimated by the kinetic equation and is shown in Fig. 3(b). At 1 K, for example, $l_{\mathrm{ph}} = 1.16$ μm is

obtained, which is two orders smaller than the sample length. The mean free path is not saturated at even less than 1 K and follows a T^{-1} dependence, which is the typical temperature dependence of glassy materials. Since the cooling rate for the QSL compounds was the same for all AFM compounds, the glasslike behavior of κ cannot be attributed to the discrepancy in the cooling process. Figure 3(c) shows the κ/T versus T plot of the $EtMe_3Sb$ salt. Even for this QSL candidate, the result of the power-law fitting indicates a small residual term $\kappa_0/T \sim 0.006$ W K^{-2} m^{-1} (the details can be found in the Supplemental Material). Note that our result for the $EtMe_3Sb$ salt agree with the data in Ref. [13].

To reproduce this temperature dependence of κ, we introduce the soft-potential model, which is a phenomenological theory usually used to describe the thermodynamic properties of glass and amorphous materials at low temperatures [37]. In this model, the temperature dependence of κ can be described as

$$\kappa = A\frac{9u^2}{1.1 + 0.7u + 3u^2} \quad \left(u = \frac{k_B T}{W}\right), \tag{1}$$

where W is the soft-potential parameter, k_B is the Boltzmann constant, and A is a temperature-independent factor that can be used as the fitting parameter. W can be calculated by $W = 1.6k_B T_{max}$, where T_{max} is the peak temperature of κ when plotted as κ/T versus T. In the case of the $EtMe_3Sb$ salt, T_{max} was 1.40 K, resulting in $W/k_B = 2.24$ K. Figure 3(c) demonstrates that the soft-potential model reproduces the temperature dependence of κ of the $EtMe_3Sb$ salt very well when the fitting parameter $A = 0.0285$. In addition, W estimated from the temperature dependence of the lattice heat capacity C_{ph} is $W/k_B = 2.39$ K (details are given in the Supplemental Material [19]). The good agreement of the W values from using different physical quantities demonstrates the validity of the soft-potential model. This suggest that the temperature dependence of κ of the $EtMe_3Sb$ salt can be understood as the classical two-level system characteristic of disordered lattice systems [38,39]. These results suggest that the contribution of spins is negligible in the $EtMe_3Sb$ salt and that the thermal conductivity is dominated by the contribution of disordered phonons.

Figure 3(d) compares the temperature dependences of the κ values of the $EtMe_3Sb$ salt (sample B) which was cooled more slowly ($\tau \sim 0.05$ K min^{-1}) with that of sample A. No thermal anomaly is observed over a wide temperature range up to 100 K, even though the ultrasound investigation of the $EtMe_3Sb$ salt detected an anomaly at 40 K corresponding to the rotation of the methyl group [40]. Furthermore, no difference in the heat transport behavior between samples A and B was observed, although the absolute value of κ for sample A was 1.5 times smaller than that for sample B. This result implies that the glassy behavior of the $EtMe_3Sb$ salt is not due to the cooling process. This also indicates that the rotation mode of the methyl group in the insulator layers is not related to the glasslike temperature dependence of κ directly, which is consistent with the result that there is no difference in the temperature dependences of κ between the $EtMe_3Sb$ salt and the $Et(CD_3)_3Sb$ salt, except for the absolute value. Because a prior x-ray diffraction study indicates no clear sign of lattice defects in the $EtMe_3Sb$ salt [14], the glasslike behavior may not originate from the initial structural disorder. To summarize the results mentioned above, the glasslike behavior of the $EtMe_3Sb$ salt is an intrinsic feature of the QSL compounds and is independent of the cooling process or disorder in the molecular arrangement.

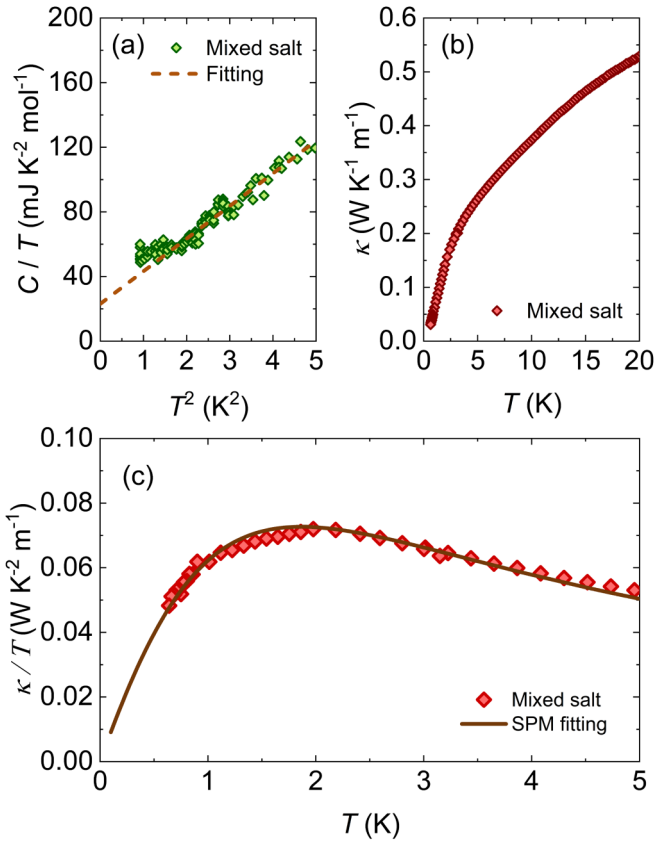

FIG. 4. (a) Temperature dependence of the heat capacity of the alloying compound of $Et_2Me_2As_{0.25}Sb_{0.75}[Pd(dmit)_2]_2$ below 5 K shown as a C/T vs T^2 plot. The orange dashed line denotes the fitting curve of $C/T = \gamma + \beta T^3$. (b) Temperature dependence of the thermal conductivity shown as aκ vs T plot. (c) κ/T vs T plot. The brown solid line is the fitting curve of the soft-potential model, where the fitting parameter $A = 0.107$.

One question is whether the glasslike behavior is specific to the $EtMe_3Sb$ salt or universal for QSL compounds in this system. Here, we presents the data for another QSL candidate without the $EtMe_3Sb^+$ ion. According to the phase diagram and chemical pressure tuning technique by Kato *et al.* [17,41,42], alloying of the cation site can be considered a rational method to survey the spin liquid state of the $Pd(dmit)_2$ system further. The cation-mixing crystal could be tuned from the AFM region to the CO region across the QSL region. The alloying compound of $Et_2Me_2Sb^+$ and $Et_2Me_2As^+$, $Et_2Me_2As_{0.25}Sb_{0.75}[Pd(dmit)_2]_2$ (hereafter referred to as the mixed salt), is such a QSL candidate ($t'/t = 0.97$). Figure 4(a) shows the temperature dependence of the heat capacity of the mixed salt. By fitting by $C/T = \gamma + \beta T^3$, we can estimate $\beta = 20.2$ mJ K^{-4} mol^{-1} and $\gamma = 23.1$ mJ K^{-2} mol^{-1}. These values are similar to those of the $EtMe_3Sb$ salt and reasonable for QSL compounds [6,7]. Figure 4(b) shows the temperature dependence of κ for the mixed salt. Even in this compound, κ

decreased monotonically with decreasing temperature, which is similar to that of pure QSL compounds. This result suggests that the glasslike temperature dependence of κ is a common feature of QSL candidates and is not unique for the $EtMe_3Sb$ salt. We note that no large residual term κ_0/T can be observed even for this compound (see the Supplemental Material [19]), although lower-temperature experiments are necessary for a decisive conclusion. As is the case for the $EtMe_3Sb$ salt, the temperature dependence of κ can be reproduced by the soft-potential model, as shown in Fig. 4(c) with the specific parameters $W/k_B = 3.08$ K and $A = 0.107$.

Our measurements demonstrate that the glassy feature of phonons is characteristic of QSL formers in $Pd(dmit)_2$-based compounds and is probably not related to extrinsic factors such as the cooling rate and disorder of molecular arrangement. However, the effect of the spin-phonon interaction may not change the temperature dependence of κ drastically. To determine the origin of the anomalous temperature dependence of κ of QSL compounds, we suppose a scenario in which the fluctuation of charge degrees of freedom confined in the $Pd(dmit)_2$ dimers changes the phonon properties into the glasslike state. In organic conductors, charge fluctuation induces modulations in periodic potentials, which can affect the phonon properties by strong electron-phonon coupling. The most typical cases are in the charge-glass system of θ-$(BEDT\text{-}TTF)_2CsZn(SCN)_4$ and rapidly cooled θ-$(BEDT\text{-}TTF)_2RbZn(SCN)_4$. They represent a steplike structure in κ and a boson peak in C owing to the strong coupling between phonons and charges [20,43]. In the case of the β'-$X[Pd(dmit)_2]_2$ series, the electronic state changes continuously from an AFM Mott insulator state to a charge-ordered state, with t'/t being the parameter, and the QSL state appears in the intermediate region in the phase diagram. It is possible that the electrons on the $Pd(dmit)_2$ dimer units fluctuate strongly in this intermediate region. In fact, a relaxorlike dielectric response is observed in the $EtMe_3Sb$ salt, suggesting the presence of disorder effects caused by the charge degrees of freedom [44]. The results of recent NMR, infrared, and Raman spectroscopy measurements suggest strong charge and lattice fluctuations in the QSL compound [45,46]. If the localized electrons work as a disorder effect on the lattice through strong charge-lattice coupling, as in θ-type compounds, the glasslike heat conduction behavior can be understood qualitatively. In addition, the dimer-Mott QSL reported has two to three times larger β values in heat capacity than those of conductive or magnetically ordered materials [6,7]. A larger low-energy lattice heat capacity is reported as a feature peculiar to $Pd(dmit)_2$-based spin liquid compounds as well as κ-type spin liquid compounds. This mechanism is consistent with the fact that κ of the $EtMe_3Sb$ salt hardly changes when a magnetic field is applied [13].

IV. CONCLUSION

In conclusion, we performed systematic measurements of the temperature dependence of the thermal conductivity of single crystals of QSL candidates and the analog compounds. The AFM compounds showed the typical temperature dependence of a crystal, whereas the QSL compounds showed a glasslike temperature dependence. It was found that the phonon thermal conductivity is systematically suppressed with increasing geometrical frustration. This result suggests that the glasslike behavior of the $EtMe_3Sb$ salt may be an intrinsic character and is not caused by extrinsic factors, such as the cooling rate or disorder in the crystals. A mechanism for the spontaneous disorder effect induced by the fluctuation of localized charges was proposed to understand the thermal transport phenomena in the $EtMe_3Sb$ salt.

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