Absence of magnetic thermal conductivity in the quantum spin liquid candidate $EtMe_3Sb[Pd(dmit)_2]_2$ — revisited

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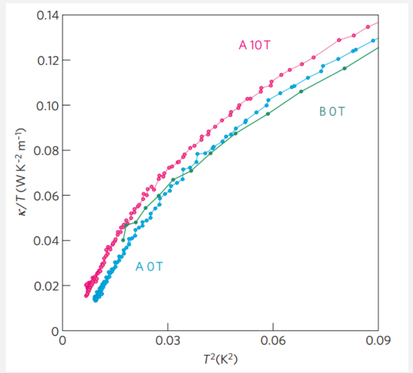
We present the ultralow-temperature specific heat and thermal conductivity measurements on triangular-lattice organic compound $EtMe_3Sb[Pd(dmit)_2]_2$ (dmit-131), which has long been considered as a gapless quantum spin liquid candidate. In specific heat measurements, a finite linear term is observed, 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 Tesla. These results are in sharp contrast to previous thermal conductivity measurements on dmit-131 [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 dmit-131 has to be reconsidered.

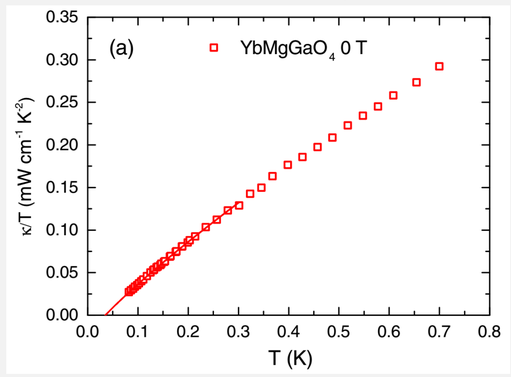
Introduction: thermal conductivity and specific heat results of triangular-lattice spin liquid candidates

κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$

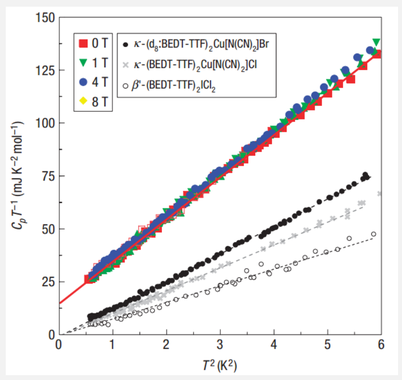
Negligible κ_0/T:
Absence of mobile gapless excitations

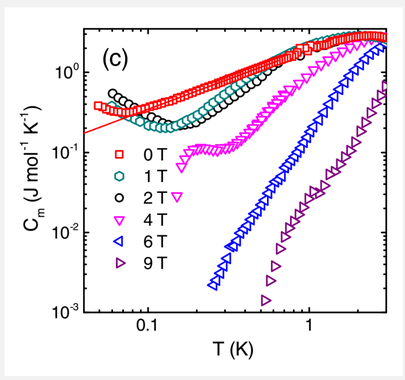


$YbMgGaO_4$



Finite γ or power law:
Gapless excitations

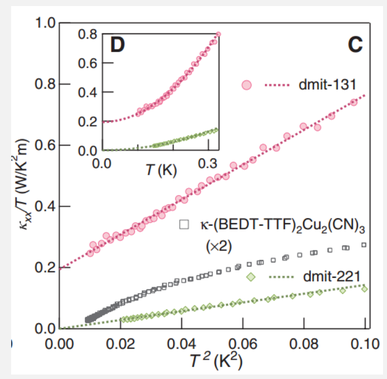




Nat. Phys. **5**, 44 (2009)
Nat. Phys. **4**, 459 (2008)
Phys. Rev. Lett. **117**, 267202 (2016)

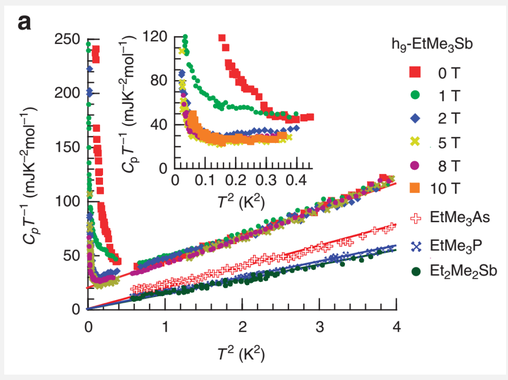
$EtMe_3Sb[Pd(dmit)_2]_2$

Finite κ_0/T:
2 mW K^{-2} cm^{-1}



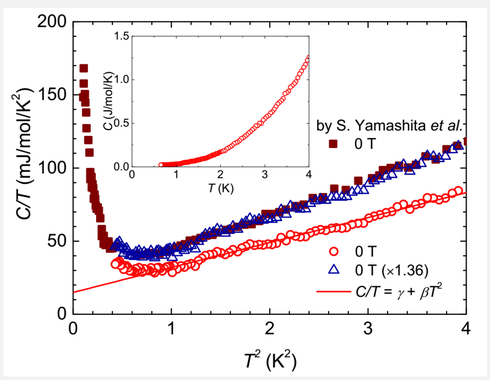
One may raise the question why dmit-131 is so different from other triangular-lattice QSL candidates.

Finite γ:
19.9 mJ K^{-2} mol^{-1}

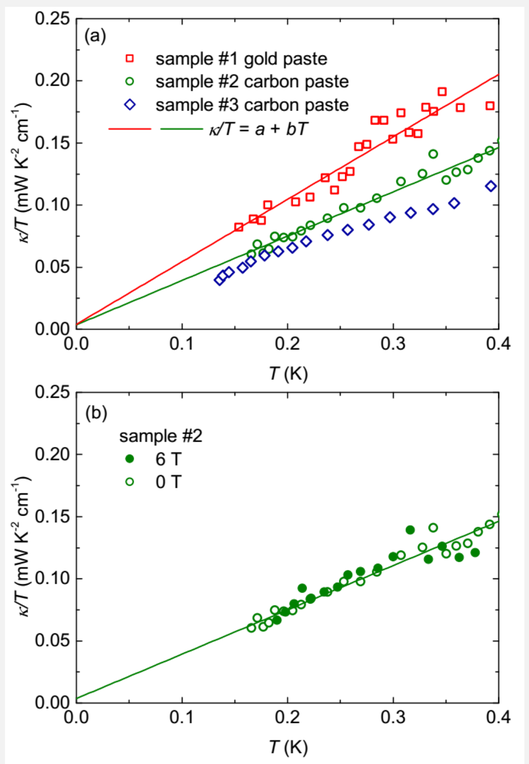


Science **328**, 1246 (2010)
Nat. Commun. **2**, 275 (2011)

Revisit the thermodynamic and transport properties of dmit-131



The previous specific heat result is well reproduced, with fitting parameters γ = 15 mJ K^{-2} mol^{-1} and β = 17 mW K^{-4} mol^{-1}.

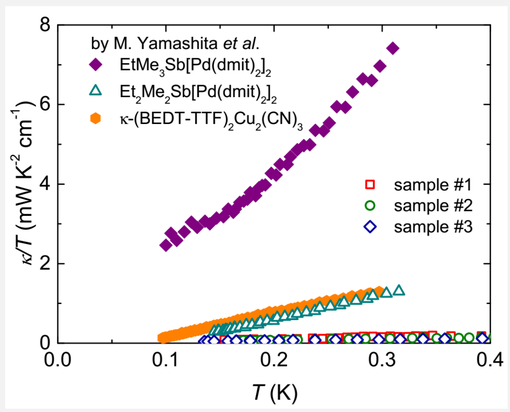


- No residual linear term κ_0/T.
- Linear or sublinear temperature dependence of κ/T.
- No change of κ in a magnetic field of 6 T.

Sharp contrast to previous thermal conductivity measurements (Science **328**, 1246 (2010)).

Inconsistent with the existence of highly mobile gapless fermionic excitations and spinon Fermi surface

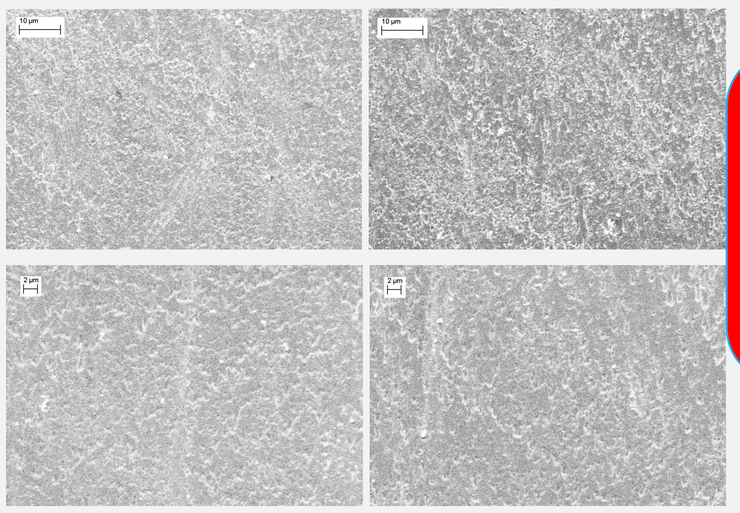
Discussion of phonon thermal conductivity



- Small absolute value and short mean free path of phonon.
- Phonons are strongly scattered by frustrated spins

Discussion of negligible κ_0/T

SEM images of sample surface after κ measurements



- No microcracks are observed.
- Absence of reproducible spinon heat transport is not due to the extrinsic factor of microcracks, and mobile excitations are intrinsically absent.

Conclusions

- A linear term in the specific heat is well reproduced, as in previous report.
- No residual linear term k_0/T is observed. A magnetic field of 6 T does not affect the thermal conductivity, and its absolute value is much smaller than the nonmagnetic reference compound.
- No magnetic thermal conductivity but only the phonon thermal conductivity in dmit-131, and the phonons are strongly scattered by the frustrated spins.
- Mobile gapless excitations are intrinsically absent in dmit-131.

J. M. Ni *et al.*, arXiv:1904.10395.