A Discrepancy in Thermal Conductivity Measurement Data of Quantum Spin Liquid β'-$EtMe_3Sb[Pd(dmit)_2]_2$

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A molecular Mott insulator β'-$EtMe_3Sb[Pd(dmit)_2]_2$ is a quantum spin liquid (QSL) candidate. In 2010, Yamashita and Matsuda reported that thermal conductivity (κ) of β'-$EtMe_3Sb[Pd(dmit)_2]_2$ is characterized by its large value and gapless behavior (a finite temperature-linear term) [1]. In 2019, however, two research groups reported that the κ values are much smaller and the temperature-linear term is vanishingly small at 0 K, which caused a serious problem concerning the ground state of QSL [2,3].

In order to explain this sharp discrepancy in the thermal conductivity measurement data, Yamashita claimed that there were two kinds of crystals (large-κ and small-κ groups) in [4] published earlier than [2,3]. Yamashita pointed out the domain formation associated with the cation disorder or the micro cracks as an origin. It should be noted that in the context of “two kinds of crystals”, the words “domain” and “micro cracks” are read as intrinsic properties that emerge in a crystal growth process or in a low-temperature phase, that is, they should be distinguished from extrinsic ones induced by improper sample handling. In response to the Yamashita’s claim, the existence of two kinds of crystals was verified using X-ray diffraction, scanning electron microscope, and electrical resistivity measurements. The conclusion is that there is only one kind of crystal [2,3].

In 2020, Yamashita and Matsuda reported that one kind of crystal gave different results in the κ measurements depending on the cooling rate [5]. In their measurements, very slow cooling (–0.4 K/h) led to larger κ values and a finite linear residual thermal conductivity. In contrast, rapid cooling (–13 K/h) resulted in smaller κ values and a vanishingly small temperature-linear term. These results suggest the existence of random scatterers introduced during the cooling process as another origin of the discrepancy. This proposal has raised a problem about effects of the newly proposed experimental parameter on other kinds of measurements.

We examined effects of the cooling rate on electrical resistivity, low-temperature crystal structure, and ^{13}C-NMR measurements and could not find any significant cooling rate dependence down to ca. 5 K [6]. Our results do not directly reject the role of the cooling rate in the κ measurements below 1 K. The cooling rate, however, engages in a process in the whole temperature region. In addition, the measurements in [1] performed with the rapid cooling rate of –10 K/h show much larger κ values than those measured with the slowest cooling rate of –0.4 K/h in [5]. This is quite puzzling and suggests that the cooling rate is not essential.

References

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