

Canted Antiferromagnetism in Novel Single-Component Molecular Conductor Consisting of Gold Dithiolene Complex

So Yokomori,^{1,2} Shun Dekura,² Tomoko Fujino,² Masatoshi Ito,² Shusaku Imajo² and Hatsumi Mori²

¹Department of Chemistry, Courage of Science, Rikkyo University, Nishi-Ikebukuro, Toshima, Tokyo 171-8501, Japan, email: s-yokomori@rikkyo.ac.jp

²The Institute for Solid State Physics, The University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8581, Japan

Metal dithiolene complexes show various physical properties in solid states due to the conjugation of π - and d -electrons (e.g., electrical conductivity and magnetism). Thus, a lot of their derivatives have been designed and synthesized, and the molecular and crystal structures, physical properties, and functionalities have been investigated to date, leading to important discoveries in chemistry and physics, such as two-dimensional π -conjugated nanosheets, single-component molecular metal/superconductor, quantum spin liquids, and Dirac electron systems [1]. Hence, the material development of new metal dithiolene complexes is significant for great progress in the fusion field of chemistry and physics.

Based on the background, we have developed novel functional molecular crystals based on metal dithiolene complexes for exploring new materials showing unique physical properties [2]. This time, a single-component molecular conductor consisting of a diethoxybenzene-substituted gold dithiolene complex (Fig. 1a) has been synthesized. This crystal is the same structure as a nickel analog [3], which is a two-dimensional herringbone structure (Fig. 1b), which is relatively rare in single-component metal-dithiolene complex crystals. The electrical conductivity at room temperature exhibited approximately 10^{-5} S/cm (Fig. 1c) and the temperature dependence of magnetic susceptibility was well fitted by the $S = 1/2$ Heisenberg model on the anisotropic triangular lattice above 20 K (Fig. 1d). Interestingly, the susceptibility showed a sudden increase at 12 K like weak ferromagnetic behavior; this is due to canted antiferromagnetic ordering. The observation of canted antiferromagnetism is very rare in molecular conductors, and its origin and mechanism will be discussed.

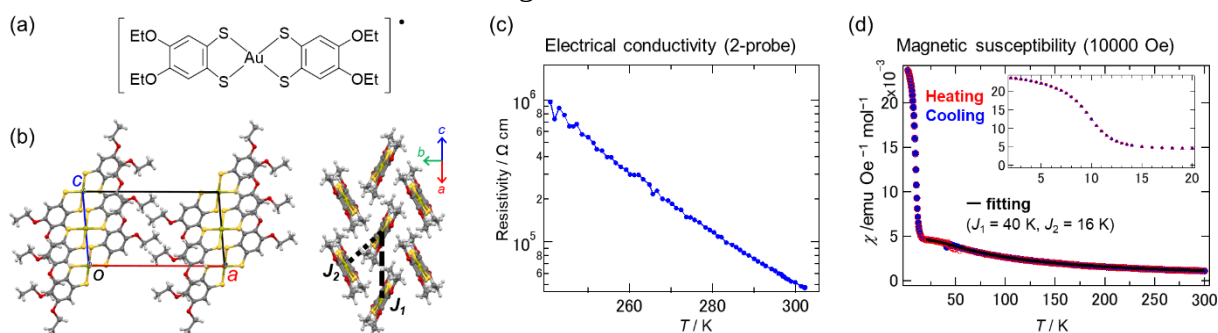


Fig. 1 (a) Chemical structure and (b) molecular arrangement of the diethoxybenzene-substituted gold dithiolene complex. Temperature dependences of (c) electrical conductivity and (d) magnetic susceptibility of the crystal.

References

- [1] T. Kambe et al., *J. Am. Chem. Soc.*, **135**, 2462 (2013); Y. L.-Gal et al., *J. Am. Chem. Soc.*, **140**, 6998 (2018); H. Cui et al., *J. Am. Chem. Soc.*, **136**, 7619 (2014); T. Itou et al., *Phys. Rev. B*, **77**, 104413 (2008); R. Kato et al., *J. Am. Chem. Soc.*, **139**, 1770 (2017) [2] S. Yokomori et al., *J. Mater. Chem. C*, **8**, 14939 (2020); S. Yokomori et al., *J. Mater. Chem. C*, **9**, 10718 (2021); H. Mori et al., *Chem. Commun.*, in press (2022), DOI: 10.1039/D1CC06826A [3] M. Ito et al., The 102nd Chemical Society of Japan Annual Meeting (2022)