Re-examination of Band Structures and Electronic Properties of Organic Dirac Electron and Related Systems

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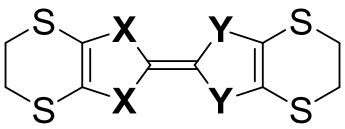
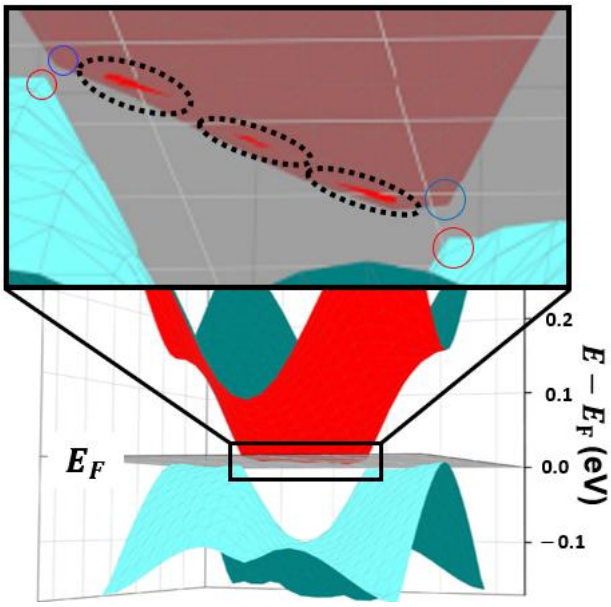


The organic conductors α-D_2I_3 (D = ET, STF, and BETS: Fig. 1) have attracted attention in terms of Dirac electron systems (DESs). Most of the studies concern α-ET_2I_3 at high pressures [1], as the appearance of DES in α-ET_2I_3 is established at ≥ 12 GPa and the samples are readily available compared to the remaining two salts. Meanwhile, at ambient pressure and room temperature, previous studies indicated that α-ET_2I_3 [1] and α-STF_2I_3 [2] are a semimetal (SM) and a zero-gap semiconductor (ZGS), respectively. α-$BETS_2I_3$ is reported to be (close to) a ZGS but with subtle differences depending on the groups [3]. Furthermore, the ground states of α-STF_2I_3 and α-$BETS_2I_3$ remain to be clarified, as the resistivities gradually increase with decreasing temperature toward T = 0 K without apparent phase transitions for both salts. In this study, we calculated the band structures using VASP (The Vienna Ab-initio Simulation Package) and re-examined the physical properties under ambient pressure to obtain consistent interpretation on the behavior of the three salts.

The striking feature of the band structures shared by the three salts lie in the flat-bottomed bands at the Fermi levels (Fig. 2), instead of Dirac cones characteristic to ZGS. In addition, The Fermi levels lie in the top HOMO (the Highest Occupied Molecular Orbital) bands in all the salts, indicating the D-I_3^- charge-transfer interactions. The band structures indicate that there are basically two kinds of electron systems in α-D_2I_3. They are fermions at the Fermi surfaces (broken ovals in Fig. 2) and thermally activated Dirac electrons near the trace of Dirac points (circles in Fig. 2). Comparing the calculated band gaps and the observed activation energies of resistivity of the three salts, the obtained band structures are quantitatively consistent with the observed electrical behaviors.



References

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[3] T. Morinari *et al.*, *J. Phys. Soc. Jpn.* **83**, 094701 (2014); D. Ohki *et al.*, *Phys. Rev. B* **102**, 235116 (2020); T. Tsumuraya *et al.*, *Eur. Phys. J. B* **94**, 17 (2021); S. Kitou *et al.*, *Phys. Rev. B* **103**, 035135 (2021).