Charge Ordering in Chiral Molecular Conductors (DM-EDT-TTF)₂ClO₄ and (DM-EDT-TTF)₂ReO₄: Raman Investigations

Iwona Olejniczak,¹ Tomasz Runka², Flavia Pop³, Narcis Avarvari³

¹ Institute of Molecular Physics, Polish Academy of Sciences, Smoluchowskiego 17, 60-179 Poznań, Poland, e-mail: olejniczak@ifmpan.poznan.pl

² Faculty of Materials Engineering and Technical Physics, Poznan University of Technology, Piotrowo 3, 60-965 Poznań, Poland

³ UMR 6200, CNRS, Laboratoire MOLTECH-Anjou, UFR Sciences, Université d'Angers, Bât. K, 2 Boulevard Lavoisier, 49045 Angers, France

[(S,S)-DM-EDT-TTF]₂ClO₄ [1], [(R,R)-DM-EDT-TTF]₂ClO₄ [1], and [(R,R)-DM-EDT-TTF]₂ReO₄ [2] [DM-EDT-TTF = dimethyl-ethylenedithio-tetrathiafulvalene] belong to a family of low-dimensional charge-transfer salts, which are characterized by chirality introduced into tetrathiafulvalene-derived radical. These layered chiral conductors crystallize in the enantiomorphic hexagonal space groups [1,2]. Metal-insulator phase transition (MI) is observed at 40 K in both the (DM-EDT-TTF)₂ClO₄ salts [1], and around 110-120 K in [(*R*,*R*)-DM-EDT-TTF]₂ReO₄ [2]. The [(*S*,*S*)-DM-EDT-TTF]₂ClO₄ and [(*R*,*R*)-DM-EDT-TTF]₂ClO₄ materials were recently shown to display an electrical magnetochiral anisotropy effect [1]. In order to provide information about the origin of the MI transition in these materials, we investigate variable-temperature Raman spectra with the focus on charge-sensitive stretching C=C vibrations of DM-EDT-TTF [3]. In our results we find clear signatures of charge-ordering (CO) in low-temperature insulating phase of [(*R*,*R*)-DM-EDT-TTF]₂ClO₄ (Fig. 1) and [(*R*,*R*)-DM-EDT-TTF]₂ReO₄.



Fig. 1. Splitting of charge-sensitive stretching C=C vibrations of the DM-EDT-TTF donor molecule observed in Raman spectra of [(R,R)-DM-EDT-TTF]₂ClO₄ (left panel) suggests the origin of the MI transition related with charge ordering; the mode labeled as v₂ together with the temperature dependence of the mode frequencies are shown on the right.

References

- [1] F. Pop et al., Nature Comm. 5, 3757 (2014).
- [2] N. Avarvari, unpublished results.
- [3] F. Pop et al., J. Am. Chem. Soc. 135, 17176 (2013).