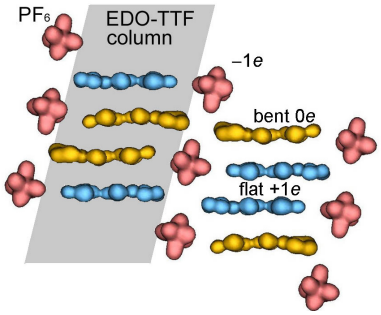
Direct observation of charge ordering in (EDO-TTF)$_2$PF$_6$, Shinobu Aoyagi,[a*] Kenichi Kato,[b] Akira Ota,[c] Hideki Yamochi,[d] Gunzi Saito,[c] Hiroyoshi Suematsu,[b] Makoto Sakata[a] and Masaki Takata[b], [a]*Department of Applied Physics, Nagoya University, Nagoya 464-8603, Japan,* [b]*SPring-8/JASRI, Kouto, Mikazuki, Sayo, Hyogo 679-5198, Japan,* [c]*Division of Chemistry, Graduate School of Science, Kyoto University, Sakyo-ku, Kyoto 606-8502, Japan, and* [d]*Research Center for Low Temperature and Materials Sciences, Kyoto University, Sakyo-ku, Kyoto 606-8502, Japan*. E-mail: aoyagi@mcr.nuap.nagoya-u.ac.jp

Keywords: charge ordering; organic conductors; charge density study

The metal–insulator (MI) transition in an organic conductor (EDO-TTF)$_2$PF$_6$ ($C_{16}H_{12}S_8O_4PF_6$) has been considered as the particular example which shows the cooperative action of Peierls distortion, charge ordering and anion ordering together with a molecular deformation. [1] The transition appears at T_{MI} = 280 K, accompanying the changeover from para- to diamagnetism. The charge ordering of EDO-TTF donor molecules in the insulating low-temperature phase has been pointed out from the comparison of the bond length and the Raman spectra. In this presentation, we report direct evidence for an ordering of (EDO-TTF)$^+$ and (EDO-TTF)^0 visualized in the (EDO-TTF)$_2$PF$_6$ charge density distributions. The charge density distributions were obtained from the synchrotron-radiation (SR) powder-diffraction data by a combination of the MEM (maximum entropy method) and the Rietveld method. [2] The SR experiment was carried out with the large Debye-Scherrer camera at SPring-8 BL02B2. The equi-charge-density surface at 260 K is shown in Figure with a level of 0.7 eÅ^{-3}. The charge on each donor molecule, coulombic interactions between PF_6^- anions and donor molecules, and the hole concentration on each sulfure atom in (EDO-TTF)$^+$ were revealed by the charge densities. The charge ordering of donor molecules was observed with a $2k_F$ periodicity ($2k_F$: nesting vector of Fermi surface). As a result, the [0,0,+1,+1,…] charge-ordering is formed along the nesting vector. Changes in the bonding during the MI transition that is dimerization of (EDO-TTF)$^+$ molecules also became evident, which can explain the existence of the insulator singlet state. In addition, a bonding between the donor molecules that suggests quasi 1D properties was found in the charge density of metallic phase.



[1] Ota, A., Yamochi, H. & Saito, G. (2002). *J. Mater. Chem.* **12**, 2600-2602.

[2] Takata, M., Nishibori, E. & Sakata, M. (2001). *Z. Kristallogr.* **216**, 71-86.