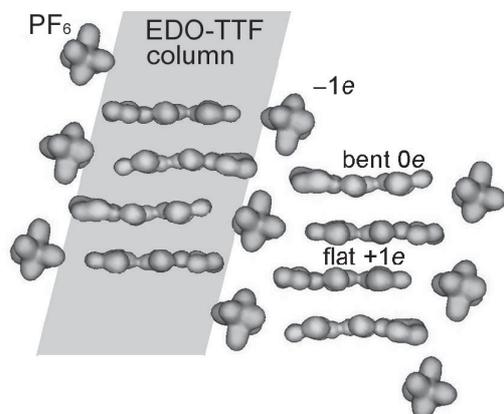


s2.m10.p1 **Direct observation of charge ordering in (EDO-TTF)₂PF₆.** Shinobu Aoyagi,^a Kenichi Kato,^b Akira Ota,^c Hideki Yamochi,^d Gunzi Saito,^c Hiroyoshi Suematsu,^b Makoto Sakata^a and Masaki Takata^b, ^a*Department of Applied Physics, Nagoya University, Nagoya 464-8603, Japan,* ^b*SPring-8/JASRI, 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)₂PF₆ (C₁₆H₁₂S₈O₄PF₆) 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)₂PF₆ 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 $0.7 e\text{\AA}^{-3}$. The charge on each donor molecule, coulombic interactions between PF₆⁻ anions and donor molecules, and the hole concentration on each sulfur 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.



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s2.m10.p2 **Intra and inter-molecular charge transfer in metal-insulator switching compound (EDO-TTF)₂-PF₆.** Nicolas Claiser^a, Brice Arnaud^a, Gwenaëlle Roger^a, Loïc Toupet^a, Thierry Roisnel^b, Akira Ota^c, Hideki Yamochi^c, Gunzi Saito^c and Philippe Rabiller^{a, a} *GMCM UMR CNRS 6626, Université de Rennes 1,* ^b *LCSIM UMR CNRS 6626, Université de Rennes 1* and ^c *Division of Chemistry, Graduate School of Science, Kyoto University, Japan.* E-mail: philippe.rabiller@univ-rennes1.fr

Keywords: Electron density; Topological analysis; Molecular switching

Among molecular switching materials, (EDO-TTF)₂-PF₆ compound presents very interesting metal-insulator phase transition where co-operative order-disorder and charge ordering take place [1,2]. In the low temperature insulating phase, the anions (PF₆⁻) are well ordered whereas two kinds of EDO-TTF molecules exist with different molecular charges: positively charged bent molecules and quasi neutral flat ones. Results of topological analysis on gridded electron density [3] derived from multipolar refinement of 40K high resolution x-ray data will be presented. Crystal effect will also be addressed based on ab-initio calculations.

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