Charge Order and its Fluctuation in α-type BEDT-TTF Charge Transfer Salts

Y. Yue[1,2]; C. Nakano[2]; K. Yamamoto[1,2]; M. Uruichi[2]; K. Yakushi[1,2]; T. Hiejima[3], and A. Kawamoto[4]
[1]*Graduate University for Advanced Studies, Okazaki, Japan*
[2] *Institute for Molecular Science, Okazaki, Japan*
[3]*Department of Nanochemistry, Tokyo Polytechnic University, Atsugi, Japan*
[4]*Department of Physics, Hokkaido University, Sapporo, Japan*
Email: yakushi@ims.ac.jp

Among a variety of BEDT-TTF charge-transfer salts, the α-type BEDT-TTF salts shows rich properties such as charge ordering, superconductivity, zero-gap semiconductor or Dirac fermion, persistent photoconduction, photoinduced phase transition, and non-linear optical response. We investigate the charge order and fluctuation of charge order in α-type BEDT-TTF salts including α′-(BEDT-TTF)$_2$IBr$_2$, α-(BEDT-TTF)$_2$I$_3$, and α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ by means of infrared, Raman, and far-infrared spectroscopy. Integrating optical conductivity, we estimated the quantity which corresponds to bandwidth, and found that the bandwidth increases in such a way as α′-(BEDT-TTF)$_2$IBr$_2$<α-(BEDT-TTF)$_2$I$_3$<α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$. α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ shows a distinct Drude response over the whole temperature range, α-(BEDT-TTF)$_2$I$_3$ does not present the well defined Drude response above the phase transition temperature. The conductivity of α-(BEDT-TTF)$_2$I$_3$ in low frequency region is almost flat meaning that the charges are strongly scattered above 136K, below which the charges are localized with a long-range order. No Drude response was found in narrow-band α'-(BEDT-TTF)$_2$IBr$_2$ over the whole temperature range. α'-(BEDT-TTF)$_2$IBr$_2$ undergoes a phase transition at around 200 K accompanying a resistivity jump by an order of magnitude. The low-temperature phase is in a charge-ordered state.[1] The high-temperature phase is a dynamically disordered state in which long-range charge order is broken.[2] The Raman-active (υ_2) and infrared-active (υ_{27}) charge-sensitive modes are split into three ascribing to nonequivalent sites in wide-band α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$, split into two ascribing to nonequivalent sites above 136 K and four ascribing to charge order below 136 K in intermediate-band α-(BEDT-TTF)$_2$I$_3$, split into two or four ascribing to charge-rich and charge-poor sites over the whole temperature range in narrow-band α'-(BEDT-TTF)$_2$IBr$_2$. The broad linewidths of these vibrational modes in high-temperature phase of α-(BEDT-TTF)$_2$I$_3$ and α'-(BEDT-TTF)$_2$IBr$_2$ may suggest the fluctuation of charge order.

[1] Y. Yue *et al*., J. Phys.: Conf. Ser. **132,** 012007 (7) (2008).
[2] Y. Yue *et al*., J. Phys. Soc. Jpn. **78**, 044701 (10) (2009).