

Infrared and Raman Studies of α -(BEDT-TTF)₂MHg(SCN)₄ with M=K and NH₄ at Low Temperature-the charge ordering fluctuation with breaking the inversion symmetry

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A family of quarter-filled organic conductors α -(BEDT-TTF)MHg(SCN)₄ with M=K and NH₄ (subsequently denote K and NH₄-salts) has been found to remarkably vary the electric structure with the kind of monocation (M). NH₄-salt has attracted considerable attention for the superconductivity at T_c=1K, whereas K-salt remain metallic down to a few mK entering a density-wave ground state below 10K which comprehends a highly controversial issue [1,2]. Recently, Dressel et al. indicated in the anomalous metallic K-salt that the charge ordering fluctuation already develop at a frequency of about 200cm⁻¹ for temperature T<200K [3]. To search independent experimental evidence for charge ordering fluctuation, we have investigated the temperature-dependent infrared (IR) and Raman spectra of K- and NH₄-salts in the edge plane.

In the optical conductivity $\sigma(\omega)$ spectra for K-salt, the charge sensitive IR active ν_{27} mode split into four bands at around 200K from the two bands observed at 1450 and 1480cm⁻¹ at 300K as shown in Figure 1. The α -phase compounds have four molecules (A, A', B, C) in the unit cell. The three of them are the crystallographically independent (A, B, C) sites, while A and A'-site are in a relationship of the inversion symmetry. From the simple symmetry consideration, the splitting of ν_{27} mode indicates the break of the inversion symmetry below 200K. Moreover, the frequency difference between A and A'-sites tend to significant extend with lowering the temperature. Consequently, K-salt exhibits to develop the charge ordering fluctuation with breaking the inversion symmetry below 200K. In contrast, NH₄-salt was found to keep the inversion symmetry down to 7.3K, although ν_{27} mode split into three band corresponding to the (A,B,C) site below 200K. We will also discuss the temperature dependent IR and Raman spectra of Rb-salt.

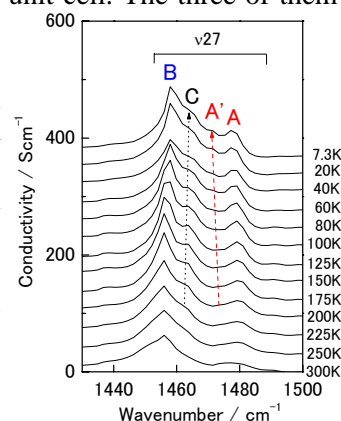


Figure 1 Temperature dependent $\sigma(\omega)$ of K-salt

[1] T. Osada et al., Phys. Rev. B. **41**,5428 (1990).

[2] J. S. Brooks et al., Phys. Rev. B **52**, 14457 (1990).

[3] M. Dressel et al., Phys. Rev. Lett., **90**, 167002 (2003)