4Pa117

Estimation of doping site by optical spectroscopy for β'-$(BEDT\text{-}TTF)_3(ZnCl_4)_{2-x}(GaCl_4)_x$

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Usually the electronic states of conductors can be controlled by two methods: electron correlation and band-filling controls. As for organic system, there were few examples whose electronic state has been controlled by the band-filling method, because it usually results the change of the crystal structure. However, Mori *et al.* have recently succeeded in the systematic study of the band-filling control in organic conductors. Among of them, the system of β'-$ET_3(MCl_4)_{2-x}(GaCl_4)_x$ (M=Zn and Co), which is isostructural to β'-$ET_3(MCl_4)_2$ (M=Mn and Zn), is studied by the optical spectroscopy in order to estimate the doping site.

The Raman and IR spectra measurements were performed on this system, not only because of the C=C stretching modes of ET molecule are very sensitive to the change of charge resided on it, but also the reflection spectrum are very powerful method to extract information for the electronic structure. For this system, the doping effect is rather complicated since there are two kinds of ET donor molecules *A* (forming conducting sheet) and *B* (sandwiched by the anions) in the pure compound holding different charge as A^{1+} and B^{2+}.

Figure 1 shows the Raman spectrum of the doped compound β'-$ET_3(ZnCl_4)_{1.42}(GaCl_4)_{0.58}$ comparing with the pure salt at both polarized parallel and perpendicular to *c*-axis. We have also carried out the Raman measurement for β'-$ET_3(CoCl_4)_{2-x}(FeCl_4)_x$ in a wide doped range of x=0.56, 0.84 and 1.24, however, the spectra of them showed almost the same pattern as β'-$ET_3(ZnCl_4)_{1.42}(GaCl_4)_{0.58}$. We found that the Raman spectrum of doping compound is different from the pure material in C=C stretching modes range. The Raman spectrum suggested that the doping in this system is more likely to be on the *A* molecule. On the other hand, the IR spectrum gives other interesting information to estimate the doping site. According to the IR spectrum from MIR to visible region, we assumed that the doping is also happened on the *B* donor molecule.

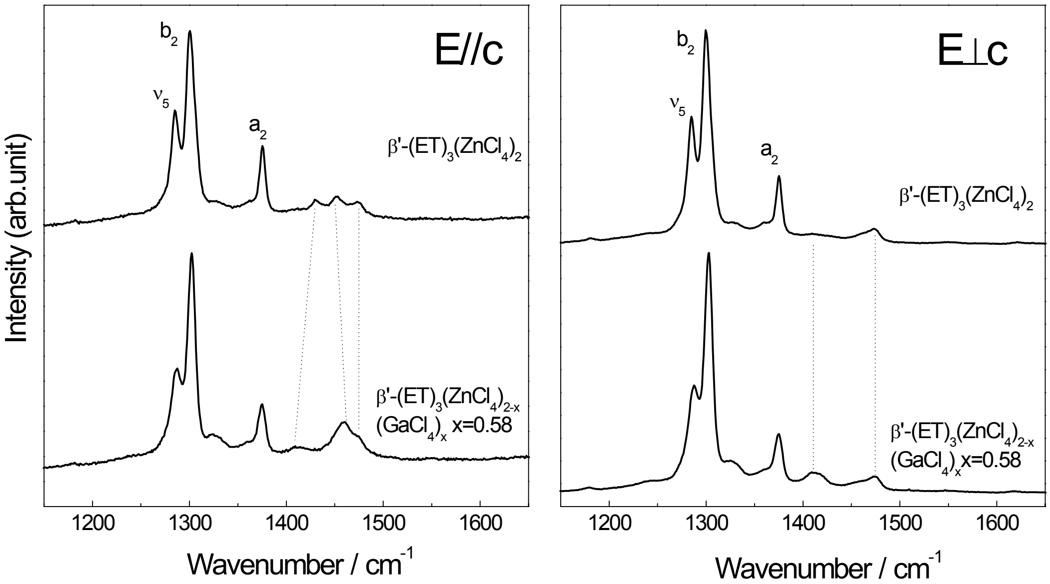

Fig.1 Raman spectra of β’-$ET_3(ZnCl_4)_{2-x}$ $(GaCl_4)_x$ x=0 (top) and x=0.58 (bottom) at polarized parallel (left) and perpendicular (right) to *c* axis