There has been a growing interest in the charge fluctuation (CF) in terms of the pairing mechanism of the superconducting (SC) transition. It is well recognized that the $\beta^*$ and $\beta$ type superconductors, whose 2D layers contain no tight dimer, exhibit CF. In contrast to this, few experimental studies have been carried out concerning the molecular superconductors whose 2D layers consist of tight dimer. We have focused our attention on the tight dimer system exhibiting the phase transition from the antiferromagnetic (AF) state to the SC phase.

We have applied vibrational spectroscopy to the AF state of EtMe$_3$As[Pd(dmit)$_2$]$_2$ ($T_N = 23$ K, $T_c = 4.3$ K at 7 kbar). Figure shows IR-reflectance and Raman spectra in the AF state. Not only the B mode but also A mode shows peak separation. The peak separation in both modes indicates that charges of dimers are inhomogeneous. The peak separation of both modes is also observed in the charge ordered (CO) state of Et$_2$Me$_2$Sb[Pd(dmit)$_2$]$_2$ [1]. In this compound, the inhomogeneous charge is static because the differences in the frequencies of two A modes and two B modes are large: 36 cm$^{-1}$ and 37 cm$^{-1}$, respectively. On the other hand, the differences in the frequencies in the present compound are $\sim$10 cm$^{-1}$ and $\sim$15 cm$^{-1}$, respectively. A small but non-zero value suggests CF: electrons of a dimer are slowly transferred to the neighboring dimers. It is suggested that the CF of the present compound is based on the inter-dimer CO state of Et$_2$Me$_2$Sb[Pd(dmit)$_2$]$_2$.

This result is in clear contrast to our previous result of triclinic-EtMe$_3$P[Pd(dmit)$_2$]$_2$ and monoclinic-EtMe$_3$P[Pd(dmit)$_2$]$_2$, the latter of which exhibits the phase transition from the nonmagnetic (NM) state to the SC phase. In these compounds, two asymmetric dimers form a symmetric tetramer. The CF of monoclinic-phase is based on the intra-dimer charge separation of triclinic-phase.

In our previous study, we have shown that the difference in the CO states between Et$_2$Me$_2$Sb[Pd(dmit)$_2$]$_2$ and triclinic-EtMe$_3$P[Pd(dmit)$_2$]$_2$ is explained from the viewpoint of the molecular orbital (MO) level characteristic of the tight dimer system [2]. It is suggested that the difference in the CF states between NM state of monoclinic-EtMe$_3$P[Pd(dmit)$_2$]$_2$ and AF state of EtMe$_3$As[Pd(dmit)$_2$]$_2$ is explained from the same viewpoint.
