Dielectric Measurements of Some \([\text{Pd(dmit)}_2]_2\) Salts Showing Charge Separation

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Most \([\text{Pd(dmit)}_2]_2\) (see Fig.1 (a)) salts are Mott insulators where one spin (\(S=1/2\)) is localized on each strong dimerized unit \([\text{Pd(dmit)}_2]_2^-\) at ambient pressure. The Cs and \(\text{Et}_2\text{Me}_2\text{Sb}\) salts undergo charge separating transitions, \(2[\text{Pd(dmit)}_2]_2^- \rightarrow [\text{Pd(dmit)}_2]_0^0 + [\text{Pd(dmit)}_2]_2^{2-}\), to a non-magnetic state \([1,2]\). Recently, we found new charge separating transitions in the salt with PyH (see Fig.1 (b)) derivative and DABCOH (Fig.1 (c)). In these salts, the charge separation undergoes with orientational ordering of cations, and the \(\text{NH} \cdots \text{S}\) hydrogen bonds are formed only with \([\text{Pd(dmit)}_2]_2^{z+}\) in the charge separated phase. Namely, the charge-separated phases are stabilized by electrostatic attractive force through hydrogen bonds. For these results, cation dynamics are obviously important for the phase transition in these salts. In this talk, we will discuss the mechanism of the charge separating phase transition as studied by dielectric relaxation and heat capacity measurements.

![Chemical structures](image)

Fig. 1. Chemical structures of (a)\([\text{Pd(dmit)}_2]_2\), (b)PyH and (c)DABCOH

![Dielectric constant vs. temperature](image)

Fig. 2. Temperature dependence of dielectric constant in (d)PyH[\([\text{Pd(dmit)}_2]_2\) and (e)Cs[\([\text{Pd(dmit)}_2]_2\)

References