Exploring Lattice Effects at the Charge Ordering Transition in (TMTTF)$_2$X

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The experimental observation of a charge-ordering (CO) transition [1] coinciding with the onset of ferroelectricity [2] in the quasi-1D conductors of the (TMTTF)$_2$X family revealed the exceptional properties of the Mott-Hubbard insulating phase in these materials. Recently we reported on the role of lattice degrees of freedom in stabilizing the charge-ordered phase in the X = PF$_6$ and AsF$_6$ salts [3]. Our results in [3] suggest that above the CO transition temperature $T_{CO}$, CO fluctuations, evident from the dielectric measurements [2] to persist up to high temperatures, cause, via S-F contacts, positional fluctuations of the anions towards their new off-center equilibrium positions. These positional fluctuations provide an effective damping of the anions’ rigid-unit modes which were made responsible for the negative thermal expansion contribution at high temperatures [3]. Upon cooling through $T_{CO}$, however, the CO becomes static, giving rise to a freezing of these modes and, as a consequence, the negative contribution in $\alpha_{c^*}$ (see figure) is no longer active. Here we review our results of high-resolution thermal expansion measurements at the CO transition on various members of the title substances including the anions X = SbF$_6$ and Br. For the X = SbF$_6$ salt, a large $\lambda$-type anomaly occurs at $T_{co} = T_P$ (the position of the resistivity minimum), which contrasts with the step-like anomaly at $T_{co}$ for PF$_6$ and AsF$_6$. The difference can be understood as a consequence of short-range Coulomb forces in the SbF$_6$ salt, where CO coincides with a metal-insulator (MI) transition, as compared to long-range forces in the AsF$_6$ and PF$_6$ salts where $T_{CO} < T_P$. For the X = Br salt, the negative contribution in $\alpha_{c^*}$ is absent, consistent with the model proposed in [3].