Charge ordering and Wigner crystal structure in the quarter-filled band Mott insulators and high pressure metals (EDT-TTF-CONMe$_2$)$_2$X, X = Br, AsF$_6$

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We report on the synthesis and application of an internal chemical pressure to effectively control, and reduce, the Mott gap in the system δ-(EDT-TTF-CONMe$_2$)$_2$X, X = Br, AsF$_6$; the detailed accounts of its Pmna, averaged room temperature structure reversible phase transition at ca. 190 K towards a low temperature *P*21/a structure; the synthesis of (^{13}C-EDT-TTF-CONMe$_2$)$_2$Br, where one carbon atom of the inner double bond is 100% 13C-enriched and single crystal ^{13}C solid state NMR spectroscopy and relaxation revealing that charge ordering occurs at room temperature and ambient pressure; the discovery of weak superstructure Bragg reflections in d-(EDT-TTF-CONMe$_2$)$_2$Br and subsequent analysis of the superstructure symmetry and refinement of an exhaustive synchrotron radiation data set; concluding to an alternation at room temperature of neutral and oxidized molecules along both the stacking a and transverse b directions in orthorhombic, non-centrosymmetric space group *P*2nn, a CO pattern compatible with ferroelectricity. The charge disproportionation and long range order crystallization of the electron gas onto every other molecular site within a three-dimensional Wigner lattice is coupled to a concerted activation-deactivation of large collections of transverse C_{sp2}–H···O hydrogen bonds and an anti-phase, static modulation of the bromide anions displacements along b. Despites the occurrence of charge ordering, the stacks remain essentially uniform, in agreement with the rich low temperature Mott physics of the system.