Recent Progress on the issue of Neutral-Ionic Transition
in Charge-Transfer Complexes

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We revisit the long standing issue, neutral-ionic transition, in terms of conductivity and magnetism in the light of electronic ferroelectricity stemming from charge transfer coupled with lattice symmetry breaking. Investigating a quasi-one-dimensional organic ferroelectric TTF-QCl$_4$ by nuclear quadrupole resonance (NQR) under pressure, we reveal a renewed pressure-temperature phase diagram, where charge-transfer instability and lattice symmetry breaking (dimerization), which occur coincidentally below 8.5 kbar, are separated at higher pressures. Above the bifurcation point, we observed a huge enhancement of electrical conductivity with highly one-dimensional nature around the charge-transfer crossover line, which is attributed to excitations of topological charges associated with neutral-ionic domain walls. In a pressure-temperature region between the charge-transfer crossover line and the dimerization transition line, NQR and nuclear magnetic resonance (NMR) experiments found that the lattice dimerization is not long-range ordered but forms a dimer liquid and that spin excitations, which violate non-magnetic ferroelectric order, are solitonic. The present results demonstrate that strong coupling between charge and lattice degrees of freedom in one dimension brings about emergent charge/spin excitations with topological natures at low energies. In the workshop, we also report on the observation of gigantic quantum critical fluctuations in the antiferroelectric DMTTF-QBr$_4$.

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