Suppression of horizontal charge ordering and coexistence of short-range charge modulations in θ-(BEDT-TTF)$_2$MM'(SCN)$_4$

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Recently, considerable attention has been focused on organic compounds θ-(BEDT-TTF)$_2$MM'(SCN)$_4$ (M=Cs, Rb or Tl, and M'=Co or Zn). The compounds are effectively two-dimensional systems at quarter filling on anisotropic triangular lattices. The dihedral angle ϕ between BEDT-TTF molecules depends on M, and it varies the anisotropy of transfer integrals between molecules. For large ϕ (M=Rb and Tl), charge ordering is observed below ~200K. While when ϕ becomes small (M=Cs), the charge ordering is suppressed, but instead two diffuse peaks are observed in X-ray scattering experiments. This indicates that short-range charge modulations with different wave vectors remain and coexist on the verge of charge ordered phase. In this coexisting region, intriguing properties have been discovered such as non-linear I-V characteristics and large magneto-resistance.

In order to clarify the origin of coexisting charge modulations, we have calculated the charge susceptibility χ_c for the extended Hubbard model at quarter filling including the nearest-neighbor Coulomb repulsion as well as electron-phonon couplings, by employing the random phase approximation. Our results show that χ_c diverges at low temperatures for large ϕ, indicating an instability of charge ordering. Whereas by decreasing ϕ, χ_c remains finite, accompanying two peaks at different wave vectors. Through the analysis of temperature dependence of these two peaks in a wide parameter range, we discuss the origin of the coexistence of charge fluctuations.