Bipolaron Density-Wave Driven By Antiferromagnetic Correlations and Frustration in Organic Superconductors*

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Theories of unconventional superconductivity (SC) in the organic charge-transfer solids (CTS) invariably focus on the spin-density wave in the $(TMTSF)_2X$ or the antiferromagnetism (AFM) in the κ-$(ET)_2X$. Many authors have presented mean-field theories of spin-fluctuation mediated SC within the Hubbard model over the past decade. Recent experiments, however, indicate that the semiconducting state proximate to SC in the CTS can be charge-ordered (CO) as well as spin-singlet, indicating clearly that AFM is not crucial for SC. We report the results of calculations on the ½-filled band Hubbard model on the anisotropic triangular lattice that indicate that there is no SC within this model for any electron-electron (e-e) interactions or anisotropy [1], in contrast to predictions based on mean-field theories.

In the second part of this talk we point out that the effective ½-filled band picture for the CTS with ½ a carrier per molecular site ignores the internal charge degrees of freedom within the dimer unit cells. Within a ¼-filled extended Hubbard Hamiltonian that includes both e-e and electron-phonon interactions on the anisotropic triangular lattice, we show that a frustration-driven transition occurs from AFM to a two-dimensional CO spin singlet state, with pairs of charge-rich sites separated by pairs of charge-poor sites [2]. We refer to this state as the Bipolaron Density-Wave or the Paired Electron Crystal. Our theory explains the low temperature spin-singlet states in θ-$(ET)_2X$ and κ-$(ET)_2Cu_2(CN)_3$. We also point out that the so-called Valence Bond Solid in $EtMe_3Z[Pd(dmit)_2]_2$ is identical to the Bipolaron Density-Wave.

In the third part of this talk we present the theory of pressure-induced SC from the Bipolaron Density-Wave. Within our theory, the bipolarons behave as negative-U sites on an effective ½-filled band triangular lattice. Pressure increases frustration further, resulting in a transition from the CO to the superconducting state [3]. The theory explains SC within the complete family of CTS within one broad umbrella mechanism. We discuss experiments within the theory.

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