Gaining Control of Molecular Ionic Nanostructures

Achievement: New two-dimensional molecular ionic structures TTF$_x$-TCNQ$_y$ (x = 1..2, y=1..13) grow on noble metal surfaces.

Significance and Impact: Multiplicity of stable molecular 2D structures, their rich phase diagram and the corresponding phase-transitions present a new opportunity for low-dimensional molecular systems with strong electron correlations.

Research Details:

- Elucidated and controlled growth of 2D molecular ionic structural phases comprising TTF and TCNQ molecules.
- Density functional theory calculations point to stability of TTF$_x$-TCNQ$_y$ 2D crystallites with varying ratio of x and y.
- Revealed dominant role of electrostatic interactions that strongly counteract segregation into unimolecular structures (x or y = 0), and strongly favors bimolecular 2D layers (which we rigorously confirmed for the 1:1 (TTF-TCNQ) phase).

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Overview:

Bulk molecular ionic solids exhibit fascinating electronic properties, including electron correlations, phase transitions and superconducting ground states. In contrast, few of these phenomena have so far been observed in low-dimensional molecular structures, including thin films, nanoparticles and molecular blends, not in the least because most of such structures have so far been composed of nearly closed-shell molecules. In this work we show the existence of the surface phase-diagram that controls the structures of tetrathiafulvalene (TTF) donor and 7,7,8,8-tetracyanoquinodimethane (TCNQ) acceptor molecules on the surfaces, and demonstrate phase-transitions that occur upon progressively increasing the density of TCNQ while keeping the surface coverage of TTF fixed. A binding motif that underlies the stable phases and infer the dominant interactions that enable the existence of the rich spectrum of surface structures is inferred.