Resonant electronic coupling enabled by small molecules in nanocrystal solids

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The future exploitation of the exceptional properties of nanocrystal (NC) thin films deposited from liquid dispersions of nanoparticles relies upon our ability to produce films with improved electrical properties by simple and inexpensive means. We show that the electronic conduction of solution-processed NC films can be strongly enhanced, without the need of postdeposition treatments, via specific molecules absorbed at the surfaces of adjacent NCs. This effect is demonstrated for Si NC films doped with a strong molecular oxidizing agent.

Si NC Field Effect Transistors fabricated with solution-processed Si NC layers activated by F_4-TCNQ exhibit the characteristic linear regime at small drain-source voltages followed by a saturation at higher voltages, minimal hysteresis, and field-effect electron mobilities about an order of magnitude higher than in the best solution-processed Si NC devices reported so far.
Density functional calculations were carried out with molecule-doped NCs. We approximated the NC films to a superlattice of packed NCs doped with molecules within its interstitialcies. To the best of our knowledge this is an original and novel approach to study the electronic structure of NC solids. It is shown that, when populated by electrons, hybrid molecule/NC states edge (and may actually resonate with) the conduction-band states of the NC solid. This provides extra electronic connectivity across the NC network as the molecules effectively flatten the electronic potential barriers for electron transfer across the otherwise vacuum-filled network interstitialcies.

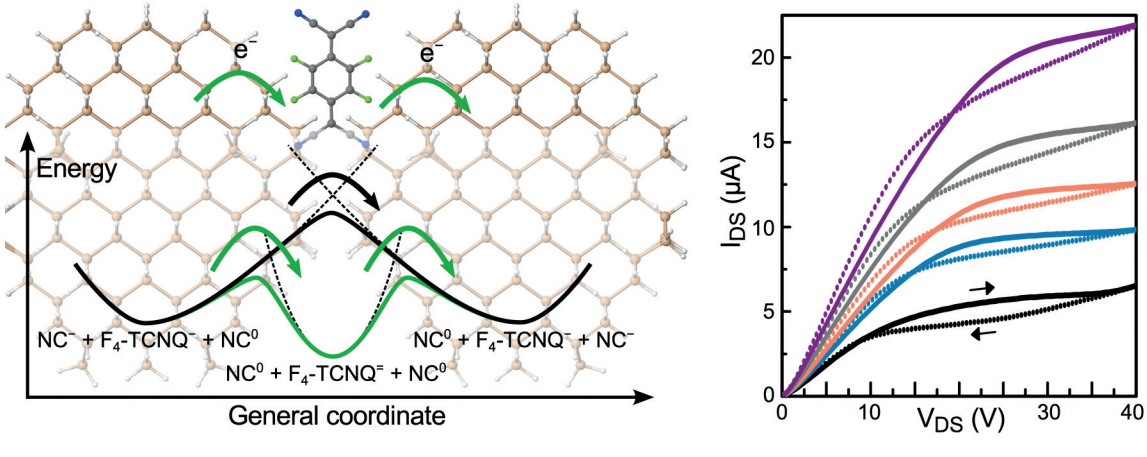

FIGURE 1
(Left panel) Schematic representation of the resonant electronic coupling enabled by small molecules in nanocrystal solids. The molecules induce new charge transfer paths (in green) responsible for the electrical activation of the films.
(Right panel) Charge transport characteristics of a printed thin film transistor fabricated with a molecule-activated nanocrystal solid.