Project Abstract

"Artificial Atoms, Molecules, and Solids: Multiple Functions and Emergent Properties"

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This presentation will describe our research in creating electronic materials from molecular and superatomic building blocks. We are designing crystalline and self-assembled systems from these building blocks. Our recent efforts have been using superatoms as the building blocks for two-dimensional materials.

One of those materials is a 2D polymerized fullerene layer. We have found that fullerenes, one of the most exotic carbon allotropes, can be polymerized to form single crystals of layered 2D fullerenes. We have synthesized a new single-crystalline 2D material by the reduction-driven covalent cross-linking of C_{60}. As a consequence of this synthetic route, these covalently linked fullerene layers are paramagnetic, with a suite of oxidation states available. The 2D structure of these crystals, bridges the gap between molecular carbon structures and extended carbon materials.

The second 2D material made from superatoms is the successfully developed spectroscopic and electrical transport techniques to probe the unprecedented properties of a 2D layered magnetic semiconductor, CrSBr. Using second harmonic generation (SHG), we find that monolayers are ferromagnetically ordered below 146 K, an observation enabled by the discovery of a large magnetic dipole SHG effect in the centrosymmetric structure. In multilayers, the ferromagnetic monolayers are coupled antiferromagnetically. Symmetry analysis establishes magnetic dipole and magnetic toroidal moments as order parameters of ferromagnetic monolayer and antiferromagnetic bilayer, respectively. The embodiment of both magnetic and semiconducting properties in this material, allows the magnetic control of interlayer electronic coupling, as manifested in tunable excitonic transitions and magnetotransport. Excitonic transitions in bilayer CrSBr and above can be drastically changed when the magnetic order is switched from layered antiferromagnetic to the field-induced ferromagnetic state, an effect attributed to the spin-allowed interlayer hybridization of electron and hole orbitals in the latter. In magnetotransport, the interlayer antiferromagnetic coupling of bilayer and thicker flakes produces a giant negative magnetoresistance response. The monolayer is drastically different, showing a maximal negative magnetoresistance at ~170 K arising from the competition between spin fluctuations and intralayer ferromagnetic ordering. Below ~30 K, a second transition ascribed to the ferromagnetic ordering of dilute magnetic defects is identified by the emergence of a large positive magnetoresistance. The magnetic coupling of the defects is mediated by carriers and we demonstrate that the magnetoresistance response can be tuned from positive to negative using a back gate to electrostatically dope CrSBr. Our work uncovers a magnetic approach to engineer electronic and excitonic effects as well as electrical transport in layered magnetic semiconductors.