We focus on developing a new class of nanoscale materials and novel strategies for the investigation of biological entities at multiple length scales, from the molecular level to complex cellular networks. Our highly flexible bottom-up nanomaterials synthesis capabilities allow us to form unique hybrid-nanomaterials that can be used in various input/output bioelectrical interfaces, i.e., bioelectrical platforms for chemical and physical sensing and actuation. We developed a breakthrough bioelectrical interface, a 3D self-rolled biosensor arrays (3D-SR-BAs) of either active field effect transistors or passive microelectrodes to measure both cardiac and neural spheroids electrophysiology in 3D. This approach enables electrophysiological investigation and monitoring of the complex signal transduction in 3D cellular assemblies toward an organ-on-an-electronic-chip (organ-on-e-chip) platform for tissue maturation investigations and development of drugs for disease treatment. Utilizing graphene, a two-dimensional (2D) atomically thin carbon allotrope, we demonstrated a new technique to simultaneously record the intracellular electrical activity of multiple excitable cells with ultra-microelectrodes that can be as small as the size as an axon ca. 2µm in size. The outstanding electrochemical properties of our hybrid-nanomaterials allowed us to develop electrical sensors and actuators, e.g., sensors to explore the brain chemistry and sensors/actuators that are deployed in a large volumetric muscle loss animal model. Finally, using the unique optical properties of nanocarbons, e.g., graphene-based hybrid-nanomaterials and 2D nanocarbides, we have formed remote, non-genetic bioelectrical interfaces with excitable cells and modulated cellular and network activity with high precision and low needed power. In summary, the exceptional synthetic control and flexible assembly of nanomaterials provide powerful tools for fundamental studies and applications in life science and open up the potential to seamlessly merge either nanomaterials-based platforms or unique nanosensor geometries and topologies with cells, fusing nonliving and living systems together.