

RESEARCH PROGRAM

Science and Engineering Abstracts for Grants Awarded in December 2021

Arizona State University

Tempe, AZ

Vladimiro Mujica, Michael R. Wasielewski, Paul S. Weiss

\$1,000,000

December 2021

Scientists from Arizona State University, Northwestern University, and the University of California, Los Angeles, will explore fundamental quantum effects in biological systems. They plan to develop and to use predictive theory and advanced spectroscopic, magnetic, and local probe techniques to elucidate the fundamental mechanisms by which molecular chirality (handedness) and spin polarization influence electron motion within biological molecules. First, they will use synthetic mirror-image (enantiomeric) pairs of DNA hairpins with additional electron donors and acceptors as part of their structures to probe chirality-dependent electron transfer. These model systems are characterized as a function of donor-acceptor distance, temperature, redox properties, and coupling to their surrounding environment to determine how chirality influences the electronic, vibrational, and spin degrees of freedom controlling electron transfer from photoexcited donors to acceptor sites as spin-coherent entangled electron-hole pairs are generated. Second, they will use magnetic substrates, nanoscale chemical patterning, and multimodal spin-polarized scanning tunneling microscopy and spectroscopies with oriented enantiomeric pairs of DNA and intercalated metals to elucidate and to quantify the molecular and interface contributions to chirality-induced spin selectivity. Since most biological molecules, including amino acids in proteins and nucleotides in RNA and DNA, are chiral, how the critical interrelated roles of spin coherence polarization, and entanglement influence electron transport within and between biological molecules will be determined. In addition to studying the unexplored roles of spin coherence in quantum biology, how it can coexist with spin polarization and how or if it can create entangled states will be addressed. The goal of this proposal is to answer these questions, which are central to and underpin the emerging field of quantum biology.

Duke University
Durham, NC
Michael Therien, David Beratan, William DeGrado
\$1,000,000
December 2021

A team of researchers from Duke University and the University of California, San Francisco plan to synthetically replicate nature's process of electron bifurcation (EB), a remarkable energy capture process that has never been achieved in a synthetic, non-natural system. In naturally occurring EB proteins move electrons from a two-electron donor to two spatially separated one-electron acceptors, sending one electron downhill to lower energy, and driving the other uphill to create a highly energized reductant. This low potential reductant is then used to catalyze green chemical processes. For example, nature uses bifurcated charges to realize highly sought-after reactions such as carbon dioxide reduction and nitrogen fixation, and to power the synthesis of life's building blocks. And yet, we have only learned how EB works in the last few years through this group's pioneering theoretical work on near-reversible natural EB systems. Proteins bind small molecule "cofactors" to direct the bifurcating charges in opposite directions by adopting shapes that fit these cofactors like a glove and dictate their function. EB thus provides a blueprint for building energy transducing nanomachines. Enabling EB by design will solve one of chemistry's grand challenges and unlock one of the most fundamental secrets of life. The researchers will combine the pinpoint accuracy of their new methods for precise design of cofactor-binding proteins, new theoretical insights on EB, with their unique ability to craft and study (with high time resolution) highly robust light-driven nanomachines. By using synthetic cofactors and proteins, they expect to achieve robustness not seen in fragile natural proteins – ultimately facilitating the design of solar energy conversion devices and green catalysis of manifold reactions unachievable in nature.

Stanford University
Palo Alto, CA
Jennifer Dionne, Tony Heinz, Felipe H. da Jornada
\$1,000,000
December 2021

Chemical manufacturing is critical for industries spanning construction, clothing, plastics, pharmaceuticals, food, and fertilizers, yet remains among the most polluting and energy demanding practices. The team's vision is to enable sustainable chemical production with atomically architected photocatalysts that precisely control optical, electronic, and molecular interactions for high-efficiency and product-selective chemistry. They will develop and deploy

excited-state quantum theory to predict and optimize the influence of atomic structure, composition, and dopants in catalytic nanoparticles on their photo-chemical properties. In addition, they will advance a revolutionary optically-coupled transmission electron microscope (OTEM) to directly test their predictions, enabling first-in-field imaging of photocatalysis *in situ* and with atomic-scale resolution. These methods will be applied to gas-phase hydrogenation reactions catalyzed by bimetallic plasmonic catalysts. By revealing the multiscale photochemical processes that span Ångstroms to centimeters and picoseconds to minutes, their combined computation and characterization will overturn the century-old empirical approach of catalyst design, and inform a new generation of sustainable, solar-driven catalytic reactions.

University of California, Davis

Davis, CA

J. Sebastian Gomez-Diaz, Joshua Hihath

\$1,000,000

December 2021

The generation and control of electromagnetic waves has helped shape the development of our society. Approaches like thermal radiation (light bulbs), thermionic emission (vacuum tubes), stimulated emission (lasers and spasers) and electroluminescence (LEDs) have been incorporated into everyday technologies. Conversely, Cherenkov radiation (CR) has been limited to niche applications such as medical imaging, dosimetry, and particle detection because CR typically requires highly energetic electrons produced by nuclear reactors, particle accelerators, or radionuclide generators. A pair of researchers will take an unprecedented approach to harness CR at long wavelengths by manipulating the coupling between drifting electrons and hyperbolic photonic modes in nanostructures made of 2D materials. This approach allows the generation and control of CR in the form of emitted photons or surface plasmons polaritons (SPPs) without the need for electron beams or exotic energy sources. Preliminary quantum-mechanical studies predict unrivaled photon emission in the terahertz (THz) and infrared (IR) bands. Their objectives are to experimentally verify this hypothesis, develop a detailed, fundamental understanding of this mechanism through a comprehensive theoretical framework able to describe the intensity, bandwidth, and operational frequencies of the electron-photon coupling, correlate experiments and theory, and explore and manipulate the properties of CR at THz/IR frequencies with the hyperbolic dispersion of nanostructures and applied bias/gate voltages. The team envisions that this approach will allow the development of miniaturized, efficient, tunable, and broadband THz and IR sources operating at room temperature, which would revolutionize the utilization of this difficult-to-access region of the spectrum for applications including communications, quantum/nano-photonics, sensing, healthcare, and beyond.

University of California, Santa Barbara
Santa Barbara, CA
Andrew Jayich
\$1,300,000
December 2021

Massive experiments, such as the Large Hadron Collider (LHC), are dedicated to finding new particles beyond the Standard Model in order to help explain basic innerworkings of our Universe. The aim of this project is to detect the signature of new physics in a tabletop experiment, to provide critical evidence of unknown particles and provide an unprecedented guide for future experiments. Rather than attempting to create these new particles in high energy collisions, a promising alternative is to search indirectly with very precise measurements of atoms or molecules to look for time symmetry violating moments of nuclei. The PI plans a transformative approach that will use just a single radioactive molecule, enabled by two sensitivity enhancements: the highly deformed radium nucleus and large, controllable molecular electric fields. Despite the experiment being intrinsically small, his analysis shows that it is possible to achieve record levels of sensitivity to time symmetry violating physics. This sensitivity will enable the detection of new physics at significantly lower cost, less time, and with a table-top scale experiment. The experiment builds upon recent advances in his group to synthesize and control radioactive molecules and work with highly sensitive, radioactive elements. The work is further supported by recent technical advances, derived from quantum information science research, that allow for record levels of control and measurement precision of molecular ions. The tabletop experiment will complement massive efforts to search for new particles and forces, but at a fraction of the cost of a collider.

University of Texas at Austin
Austin, TX
Ginny Catania, Timothy Bartholomaus, John Goff, Sean Gulick, John Jaeger
\$1,500,000
December 2021

A team of researchers from the University of Texas at Austin, the University of Idaho, and the University of Florida aims to quantify the rates of processes contributing to sediment build-up at the termini of outlet glaciers (producing moraines) to improve predictions of ice sheet response to climate. Their observations will determine the degree to which moraines can be built fast enough to form support that can offset the retreat of glaciers initiated by climate perturbations and will test the idea that moraine building is responsible for heterogeneous rates of glacier

retreat and larger steady-state glaciers than would be possible in the absence of sediment. Importantly, this project makes possible model improvements in the field of coupled ice-sediment dynamics that future sea level estimates rely on. This will be accomplished with first-of-their-kind observations and sampling at active terminal moraines in Greenland using a remotely operated vehicle (ROV) called Nereid Under Ice (NUI). This is a vehicle that is purpose-built for surveying and sampling in deep, ice marginal environments and will obtain unprecedented geological, geophysical, and oceanographic measurements at the critically under-observed ice-sediment-ocean interface. The team's observations across three glaciers in Greenland, each with distinct histories and geometries, are expected to reveal the rates and distribution of processes contributing to moraine-building, but importantly also provide ice dynamic controls. Obtaining this detailed, mechanistic understanding of moraine-building will allow far more accurate sea-level-projecting models of ice sheets to be built, thus making society more resilient to climate change.