## INTRODUCTION TO OUR RESEARCH

**WHAT IS QUANTUM INFORMATION SCIENCE (QIS)?** QIS is both new and exciting. In the classical world, Newtonian physics dominates through application of the equations of motion. As devices continue to shrink in size, quantum theory takes over. A "classical" bit of information exists in two states, "0" and "1". Quantum mechanics allows for *superposition states*, such as a linear combination of "0" and "1" and *entanglement* where if "0" and "1" are entangled a measurement of one will be directly correlated with the other. QIS research exploits the inherent properties of quantum superposition and entanglement of *e.g.*, electron and nuclear spins (qubits) to transform future technologies that include quantum sensing, quantum computing, quantum communications, quantum metrology and quantum cryptography. Two key topics recently highlighted in the *BES Roundtable on Opportunities for Basic Research for Next-Generation Quantum Systems* and further elaborated on in the more recent DOE Workshop on *Exploiting Photons and Spins in Chemical Systems for Quantum Information Science* describe (a) increased need for new quantum-coherent systems having unprecedented functionality, and (b) the need for enhanced creation and control of coherence in quantum systems.

**WHAT ARE WE DOING?** Our research is geared toward developing the ability to control excited state (ES) electronic and magnetic properties in new molecular multi-spin systems. This work is critical to developing new photocatalysts, solar energy conversion devices, and spintronic and photonic materials. We employ a combined spectroscopic approach (multifrequency electron paramagnetic resonance (EPR), magnetic circular dichroism (MCD), electronic absorption, X-ray absorption (XAS), resonance Raman (rR), emission, and time-resolved spectroscopies coupled with extended X-ray absorption fine structure (EXAFS)) to studying ES spin manipulation. These efforts play an important role in QIS, which seeks to take advantage of both superposition and entanglement. Superposition and entanglement are being used to revolutionize computing and sensing technologies. Our approach to molecular QIS systems is aligned with these *Roundtable/Workshop* topics. In collaboration with the Shultz group and NC State University, we design and synthesize bespoke radical-elaborated compounds which possess ES magnetic exchange interactions that control ES lifetimes and ES electron spin polarizations. Our primary focus is to design and spectroscopically probe novel molecular systems that retain the memory of excited state spin polarizations in their recovered ground states. These excited states are created by optical pumping using visible light, and lead to ultrafast generation of exchange-coupled spin qubits and exchange-modulated electron spin polarizations. Strong spin polarization in the GS requires that ES $\rightarrow$ GS relaxation be orders of magnitude faster than GS longitudinal and transverse relaxation times. This key criterion has been realized in our novel molecular systems. Thus, the photophysical properties of our molecules display tremendous promise for understanding how to address, control and manipulate molecular spin qubits, furthering our understanding of their spin dynamics and communication between qubits. Our *long-term goal* is to develop novel molecular systems that can be optically initiated to enable pairwise magnetic exchange mediated multi-qubit entanglement, facilitate large ground state spin qubit polarizations, and develop spatial and temporal control of spin qubits. We employ the use of modular synthetic design principles that we have developed in order to increase our understanding of how charge-transfer chromophores couple with persistent spin-based substituents to produce robust entanglement, modulate ground state spin polarizations, and promote spin coherence lifetimes that are markedly longer than the initial spin initiation process.