*Abstract: We will describe recent work from our laboratory that focuses on using photo-driven processes to generate entangled electron spins in molecular materials targeting quantum information applications. We will show how ultrafast photogeneration of electron-hole pairs within covalent electron donor-acceptor systems results in two entangled spins having sufficiently long coherence times to perform a variety of operations relevant to quantum information. We will also describe how molecular chirality influences the spin dynamics of photogenerated electron-hole pairs in donor-chiral bridge-acceptor molecules. Finally, we will show how molecular diradicals can serve as versatile surrogates for nitrogen vacancy defect centers in diamond.*