Quantum information promises advances in computation not seen since the dawn of the digital computer, but quantum logic operations occur in fragile nonequilibrium states that rapidly and irreversibly collapse to Boltzmann equilibrium. These dynamics result in a “tyranny of temperature” that makes quantum circuits noisy at temperatures higher than a few Kelvin, severely limiting the fidelity of quantum computations on most hardware. Chemistry has a critical role to play in the advancement of quantum materials by discovering and articulating molecular structure/quantum function relationships, and here I will describe how carefully designed molecules can exhibit long-lived spin coherence in specifically initialized quantum state registers at temperatures far exceeding those encountered in conventional quantum hardware. I will describe our theoretical and computational work into repurposing singlet fission, a rare photoconversion process originally envisioned as a way of improving solar cell efficiencies, to applications in quantum information. I will describe how to exploit molecular symmetries to generate strong selection rules for the nonadiabatic interconversion of the maximally entangled biexciton state produced through singlet fission to optically pump a specific magnetic sublevel. These principles come to life in our theoretical analysis on time-resolved electron paramagnetic resonance experiments of solution-phase molecular dimers and crystals, where the spin coherence time of the quintet can be long-lived according to diVincenzo’s criterion, near liquid nitrogen temperatures.