Understanding electron spin decoherence

Organic radicals and paramagnetic transition metal complexes can serve as molecular electron spin qubits for potential applications in quantum information science and technology. An important prerequisite for these applications is a long coherence lifetime. We present an experimental and theoretical investigation into the decoherence mechanism of nitroxide and other organic radicals, who possess decoherence times (phase memory times) on the order of 5 μs below 50 K in frozen protonated aqueous solutions, and more than an order of magnitude longer in fully deuterated environments. The experiments reveal that the decoherence time depends critically on the bulk proton concentration and on local spatial correlations between protons. Fully coherent many-body quantum dynamics simulations of the electron spin and about 1000 neighbouring hydrogen nuclei (protons and deuterons) quantitatively reproduce the measured coherence decays. This demonstrates that the electron spin decoherence is driven by many small clusters of magnetic nuclei, in particular geminal proton pairs (CH2 or OH2), that interact among themselves and with the electron spin.