Spin Dynamics of Molecular Electron Qubits with Pulsed EPR

Krishnendu Kundu

National High Magnetic Field Laboratory, Florida, USA

Electron spins in molecules have been proposed as potential qubits in quantum computers. But quantumness (coherence) of a spin is very fragile and it is susceptible to magnetic fluctuation present in the surroundings. However, Clock Transitions (CTs) at avoided level crossings between a pair of spin states offer a successful strategy to protect coherence against magnetic noise. More precisely, quadratic energy dependence on the magnetic field at the avoided level crossing protects the electron spin from fluctuations that couple linearly to the spin. I shall discuss two lanthanide systems to explain spin dynamics at CT.

Nuclear spins often act as the (inevitable) source of magnetic fluctuations for electron spins. The first system is a molecular holmium nanomagnet (HoW_{10}), which has crystal-field originated CTs, and has been considered as a benchmark example to understand the origin of insensitivity of electrons to nuclei at CTs. Utilizing pulsed EPR, we have demonstrated that hyperfine interactions with nuclear spins effectively vanish exactly at a CT, which eventually leads to an enhanced coherence. A many-body quantum simulation has been performed to understand the phenomenon in more detail.

The problem with crystal-field originated CTs is strong orientation dependence. The second example is based on the search for an isotropic CT system. We found a series of lanthanides (^{175}Lu) complexes, where the spin-bearing orbital extensively mixes with an s-orbital and enhances the electron-nuclear Fermi contact interaction. As Fermi interaction is a scalar quantity it generates a gigantic (nearly) isotropic hyperfine interaction (A_{iso} = 3.5 GHz or 1230 G), which, in turn, generates a 9.2 GHz hyperfine clock transition. Pulsed EPR studies reveal an order of magnitude increase in coherence time at the clock transition.

I shall also discuss a recent hardware development in implementing frequency-swept (chirp) pulses to enable pulsed EPR to perform in the Fourier transform method. The discussion will eventually include the potential usage of chirp pulses on electron qubits for the preparation of the spin states and enhancing their signal.