13C spin hyperpolarization achieved by combined incoherent and coherent processes in NV-center diamonds

G.A. Alvarez[1], C.O. Bretschneider[1], R. Fischer[2] and L. Frydman[1]
[1] Department of Chemical Physics, Weizmann Institute of Science, Israel
[2] Department of Physics, Technion, Israel Institute of Technology, Israel

Nuclear spins are candidates for storing quantum information due to their long decoherence times; they are also of fundamental interest in medicine, biology and chemistry thanks to their central roles in nuclear magnetic resonance (NMR) and magnetic resonance imaging (MRI). These uses come from the nuclear spins's weak interactions, making the energies they involve small but also their observation very challenging. In particular, at room temperatures, their very low polarization leads to low NMR/MRI signal to noise ratios. Dynamical nuclear polarization (DNP) bypasses these limitations by transferring the electron polarization to the nuclear spins. Electron spins in nitrogen vacancy (NV) centers in diamonds can be almost fully polarized with laser irradiation. DNP methods on NV diamonds have so far relied on on-resonance, coherent transfers, which demand an extreme fine tuning of the experimental's parameters, i.e. magnetic field strengths, alignment of the diamond with respect to the magnetic field, etc.

We present a novel approach for hyperpolarizing nuclear spins, based on the combined action of non-commuting coherent and incoherent processes. Besides its conceptual novelty, the strength of this approach rests in that it allows for off-resonance polarization transfers, relaxing the degree of control that needs to be exercised on the system. Ingredients of this new approach include microwave (MW) transitions addressing solely the 0 $\rightarrow$-1 (or +1) states of the S=1 electron center. These selective transitions, together with the dynamic laser hyperpolarization of the electron spin 0 state, can produce a periodic imbalance of the nuclear spin population. This is due to the asymmetry of the hyperfine interaction between the electron and nuclear spins, depending on whether the electron state is 0 or ±1. While this is not in and of itself sufficient to produce a net buildup of nuclear polarization, the iterative interferences between these MW-induced coherent transitions and relaxation-driven incoherent T1 and T2-like decays, increases the population of either a "dark" or a "bright" nuclear spin state, depending of the relation between the Rabi field, the hyperfine interaction and the relaxation time. The net results is a hyperpolarization of the nuclear spin, robust vis-à-vis many different conditions for the Hamiltonian parameter of the system, relaxing the fine-tuning requirements for the experimental setup. Laser-triggered, NMR-detected experiments exploring this effect and demonstrating the nuclear ensemble hyperpolarization are presented. This method provides new perspective for NMR/MRI applications and nuclear state initialization for quantum information processing.