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Dynamic Nuclear Polarization Breaking out of the spin diffusion barrier?

Dynamic nuclear polarization (DNP) has proved to be a powerful and versatile means to overcome the intrinsically low sensitivity of NMR[1]. A great challenge for DNP at present is to achieve high spin polarization in shorter times, which requires understanding spin polarization transfers but also spin diffusion dynamics[2]. Spin diffusion is indeed of major importance nearby the free electrons, precisely where DNP is supposed to be most efficient, but unfortunately paramagnetic shifts are said to prevent nuclei from sharing polarization with the rest of the sample. As a consequence, the so-called diffusion barrier is ubiquitous in the literature on DNP and MAS-DNP[3].
Using simple DNP and 1H pulsed NMR experiments, we have witnessed the existence of an invisible polarization spin reservoir that is able to replenish significant polarization to the observable proton spin reservoir, after full saturation and switching off the microwaves.
A two-reservoir model can efficiently describe the experimental data and allows determination of the polarization exchange rate between the visible and hidden reservoirs, as well as their intrinsic relaxation and build-up rates. Relaxation were found to be significantly lower at 1.3 K than 3.8 K, as expected. Intriguingly, the exchange rate was also found to have a similar dependence, suggesting a phonon mediated mechanism.
Several possible explanations are proposed to account for the existence of this hidden reservoir: 1H Zeeman order nearby electrons, 1H-electron dipolar order and 2H Zeeman order cross-relaxing to 1H. The relevance of these possible contributions will be discussed, and supported by further experiments.
We believe this finding will shed light on one of the most important, and misunderstood, fundamental mechanism of DNP.

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Primary authors: Mr CHAPPUIS, Quentin (Univ Lyon. Université Claude Bernard Lyon 1, ENS de Lyon, FRE 2034, CRMN, 5 Rue de la Doua, 69100 Villeurbanne, France); Dr COUSIN, Samuel (Université Claude Bernard Lyon 1); Dr ELLIOTT, Stuart James (Univ Lyon. Université Claude Bernard Lyon 1, ENS de Lyon, FRE 2034, CRMN, 5 Rue de la Doua, 69100 Villeurbanne, France); Dr CALA, Olivier (CRMN); Prof. JANNIN, Sami (Université Claude Bernard Lyon 1)

Presenter: Mr CHAPPUIS, Quentin (Univ Lyon. Université Claude Bernard Lyon 1, ENS de Lyon, FRE 2034, CRMN, 5 Rue de la Doua, 69100 Villeurbanne, France)

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