Dynamic Nuclear Polarization and Nuclear Spin Diffusion in Nanoscale Si Particles

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We investigate dynamic nuclear polarization (DNP) of ^{29}Si nuclei in spherical Si nanoparticles (NPs) and irregular shaped porous Si (PSi) NPs both possessing Si crystalline cores in the range of $25-50$ nm. DNP relies on the transfer of spin angular momentum from highly polarized, rapidly relaxing unbound electron spins to slowly relaxing, lowly polarized nuclear spins. A near-resonant microwave radiation then transfers the high electronic polarization to hyperfine coupled nuclear spins and the nuclear spin diffusion spreads the polarization from near-surface ^{29}Si spins into the crystalline cores.

Spherical Si NPs were fabricated from silane gas using bottom-up laser gas phase synthesis. PSi NPs were prepared via top-down low-load metal assisted catalytic etching [1] from electronics grade Si wafers with different dopants (boron or phosphorus) and doping densities (from $4 \cdot 10^{18}$ cm^{-3} to nominally undoped). Electron spins in the (P)Si NPs originated from the fixed paramagnetic dangling-bond type electronic centers (P_b centers), which were naturally formed in the Si/SiO_2 interface during the oxidation of crystalline cores. Performing DNP at two magnetic fields (3.4 T and 7 T) and two temperatures (1.4 K and 3.4 K) we achieved a bulk nuclear polarization of about 6 % and room temperature relaxation time of 75 ± 3 min in lightly boron doped PSi NPs.

Several conclusions on the DNP process in (P)Si NPs can be made based on the obtained data. Triple (electron-electron-nuclear) spin flips seem to be responsible for the initial hyperpolarization transfer from electron to nuclear spins. The central ^{29}Si nuclear spin of a P_b center is rapidly polarized upon microwave radiation (Fig. 1a) owing to its large hyperfine coupling of $210-417$ MHz. Microwave irradiation itself induces additional relaxation [2]. Nuclear spin diffusion in the crystalline cores (bulk) is fast and equalizes the nuclear polarization in few tens of seconds inside 50 nm Si NPs [3]. Therefore, nuclear spin diffusion does not explain the long relaxation times. The rate limiting step in low temperature DNP and room temperature relaxation is then the nuclear polarization transfer across the spin diffusion barrier — a barrier between the strongly hyperfine coupled central ^{29}Si nuclei and ^{29}Si in the bulk (Fig. 1b). Possible nuclear spin diffusion dependence on doping type and density is unimportant except possibly for highly donor doped Si beyond the metal-to-insulator transition.

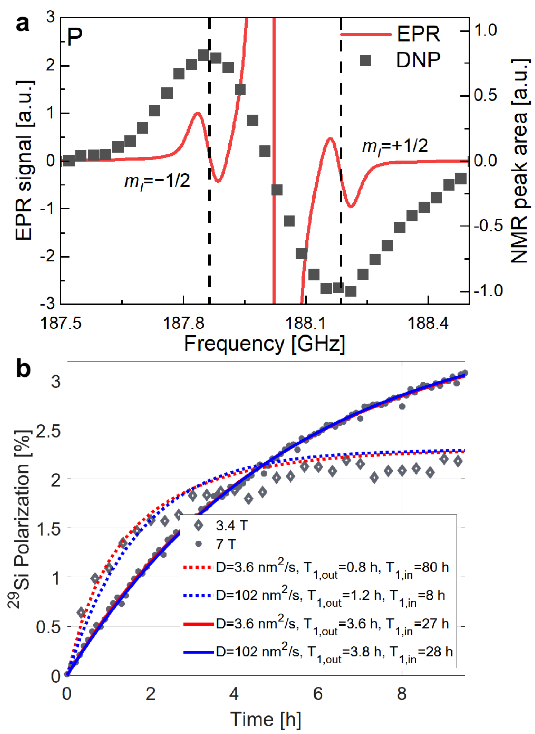

Fig. 1. (a) Overlay of an electron paramagnetic resonance (EPR) and DNP spectra at 6.7 T and 1.4 K for the PSi NP sample with $5 \cdot 10^{14}$ cm^{-3} B doping. (b) Independence of polarization buildup dynamics from the nuclear spin diffusion coefficient in 20 nm NPs at 3.4 and 7 T.

References

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