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Magnetic-field-induced delocalization in hybrid electron-nuclear spin ensembles

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We use field-cycling-assisted dynamic nuclear polarization to demonstrate magnetic-field-dependent activation of nuclear spin transport from otherwise isolated strongly-hyperfine-coupled sites. With the help of a toy model comprising electron and nuclear spins, we recast our observations in terms of a dynamic phase diagram featuring zones of active or forbidden nuclear spin current separated by boundaries defined by the interplay between spin Zeeman, dipolar, and hyperfine couplings. Analysis of the polarization transport as a function of the driving field reveals the presence of many-body excitations stemming from the hybrid electron-nuclear nature of the system. These findings could prove relevant in applications to spin-based quantum information processing and nanoscale sensing.

Keywords: Nuclear spin diffusion | Dynamic nuclear polarization | Nitrogen-vacancy centers | Thermalization | Many-body localization

The connection between statistical mechanics and the dynamics of isolated many-body quantum systems is typically formulated in terms of the eigenvalue thermalization hypothesis[1-5] (ETH), the idea that complexity in interacting systems prompts ergodicity at the quantum level. Disorder and quantum interference can stymie thermalization, often leading to regimes of sub-diffusive dynamics or suppressed transport, as already identified in a broad class of systems ranging from acoustic and optical waves to cold atomic gases[6-8]. The breakdown of ergodicity can extend even to strongly interacting quantum systems, a regime known as many-body localization[9-11] (MBL). While most MBL experiments thus far have focused on isolated arrays of trapped atoms[12,13], recent theoretical work argued that the interplay between diffusion and localization also influences the out-of-equilibrium dynamics of driven open systems[14,15]. In particular, these studies built on the extensive body of work on dynamic nuclear polarization[16,17] (DNP) to show that the concept of spin temperature is directly connected (namely, relies on) quantum ergodicity and ETH.

Indeed, ensembles of electron and nuclear spins in solids provide a practical experimental platform to investigate thermalization because disorder and long-range interactions compete in ways that can be exposed using alternative spin control techniques. For example, recent experimental work studied spin depolarization in ensembles of nitrogen-vacancy (NV) centers in diamond and revealed sub-exponential, disorder-dependent relaxation associated with critical thermalization dynamics[18,19]. Along similar lines, dynamic nuclear polarization of carbon spins in diamond was exploited to expose electron-spin-mediated nuclear spin diffusion exceeding the value expected for naturally abundant ^{13}C spins by nearly two orders of magnitude[20].

Here, we resort to nuclear spins in diamond to demonstrate control over the localization/delocalization dynamics of hyperfine-coupled carbons upon variation of the applied magnetic field. We formally capture our observations by considering a model electron-nuclear spin chain featuring magnetic-field-dependent spin transport. Further, we show the dynamics at play can be cast in terms of well-defined dynamic phases that can be accessed by tuning the magnetic field strength and paramagnetic content. The spin state hybridization emerging from the intimate connection between electron and nuclear spins gives rise to otherwise forbidden low-frequency transitions, whose presence underlies the system's singular spectral response to RF excitation of variable amplitude.

In our experiments, we dynamically polarize and probe ^{13}C spins in a [100] diamond crystal ($3\times3\times0.3$ mm^3) grown in a high-pressure/high-temperature chamber (HPHT). The system is engineered to host a large (~10 ppm) concentration of NV centers, spin-1 paramagnetic defects that polarize efficiently under green illumination. Coexisting with the NVs is a more abundant group of P1 centers (~50 ppm), spin-1/2 defects formed by substitutional nitrogen atoms. We tune an externally applied magnetic field B in and out of the 'energy