**Supplementary Information**

Electron induced nanoscale nuclear spin relaxation probed by hyperpolarization injection

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Fig. S1. $T_1$ measurements. Sample $T_1$ at low-field (36mT, blue) and high-field (7T, red) at $\tau=60s$. Points are data and solid lines are monoexponential fits. Dashed line denotes $1/e$, and intersections give a low-field $T_1$ of 283$\pm$2s and a high field $T_1$ of 1520$\pm$20s.

**I. $T_1$ MEASUREMENTS AT LOW AND HIGH FIELD**

**Fig. S2.** Polarization buildup with hyperpolarization period $\tau$. Panel shows the unnormalized data corresponding to Fig. 3A of the main paper. The different curves are the $^{13}C$ pulsed spin-lock decays obtained under varying hyperpolarization time $\tau$ (see colorbar). This data is also shown as a movie in Ref. [40].

The eye to track the slowing relaxation dynamics with increasing $\tau$. **Fig. S2** shows an unnormalized plot of Fig. 3, from where the polarization buildup dynamics as a function of $\tau$ can be extracted.

**II. MOVIES SHOWING DATA IN FIG. 2 AND FIG. 3**

As a complement to the graphs in the main paper, we present movies corresponding to the data in Fig. 2A and Fig. 3A on Youtube (found at Refs. [38, 39]). These movies show clearly the progressive slowing down of the decay profiles upon increasing the hyperpolarization time $\tau$ in both representations. The gray lines here show fitted stretched exponential lines corresponding to the previous data for clarity, allowing a guide to

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**Fig. S3 describes the two relaxation times present in the experiments in the main paper. In the system of $^{13}C$ nuclear spins we consider, free induction decay lifetime $T_1^*$ $\approx$1.5ms, predominately due to strong dipolar interactions between individual spins (Fig. S3A). Under Floquet control (described by Fig. S3B(i)), however, $T_1^* = 90.9s$, an extension of more than 60,000x. In fact, the signal remains substantial even at $\sim$600s, as shown in Fig. S3B (here hyperpolarization time $\tau = 120s$). This dramatic increase in lifetime is arranged for through spin-locking to a conserved axis (x) which is arranged collinear with the initial magnetization._

**Fig. S3C** shows that the $T_1^*$ lifetime is dependent on the spin-lock pulse sequence parameters, namely the driving frequency. Here, $I\rho$ is used as a convenient metric for describing the fre-
Fig. S3. Relaxation lifetimes under Floquet control. (A) Free induction decay of $^{13}$C nuclei in $T^*_2 \approx 1.5$ ms due to strong dipolar coupling. (B) Lifetime extension due to spin-locking to $T'_2 = 90.9$s. Floquet driving consists of a train of $\phi$ pulses separated by acquisition periods (inset). Here $t_{\text{acq}}=2\mu$s, $t_p=40\mu$s, and $\rho=99.28\mu$s. (C) Spin-lock lifetime dependence on drive frequency. Traces shows an increase in lifetime with decreasing $J_\rho$. Here, $J$ is the median dipolar coupling strength and $\rho$ (the interpulse spacing in the Floquet drive) is varied. Figure adapted from Ref. [23].

IV. ASSUMPTIONS MADE FOR THEORETICAL MODEL IN FIG. 4

In what follows, we summarize the main assumptions made for the theoretical model utilized in this work and provide justifications of their relevance:

1. **Uniform nuclear spin density**: we have assumed here that the $^{13}$C nuclei are uniformly distributed in the lattice with a high density, hence allowing Eq. (1) to be written as a continuous differential equation with respect to $r$. This is a good approximation in our experiments because they constitute an ensemble average over $>10^8$ NV-$^{13}$C systems; each with a central NV defect surrounded by $\sim 10^4$ $^{13}$C nuclei. Any microscopic features rising from the discrete arrangement of the spins are washed out by the ensemble average.

2. **Purely radial polarization distribution**: we have assumed that the polarization profile is isotropic in space and only depends on the radial position $r$ from the NV center. This allows the diffusion equation in Eq. (1) to be written for a single variable $r$. While we expect some variability in the rate of hyperpolarization injection from the NV center based on angle theta (due to variation in the hyperfine coupling), any differences in polarization within a radial shell are rapidly homogenized by $^{13}$C spin diffusion (strong for nuclei in the same shell because they are at identical $r$).

3. **Absence of spin diffusion barrier**: in our model we have assumed spin diffusion between all $^{13}$C nuclei ignoring the presence of a spin diffusion barrier. This simplifying assumption is valid because the finite measurement bandwidth in our experiment makes it feasible to only measure nuclei that are weakly hyperfine coupled, i.e. with $0 < |A_{zz}| < 30$kHz. This region can be considered close to the boundary of the spin diffusion barrier, and the number of spins in this region affected by the barrier (with their diffusion suppressed) are relatively very few.

4. **Validity of the spin diffusion model**: we have assumed pure hydrodynamic diffusion in a Bloembergen-like model [44]. This assumes that the polarization evolution can be captured by Fick’s law. This is a good approximation given the spin numbers we consider; every NV center has $\sim 10^4$ $^{13}$C nuclei surrounding it, and at the distance scales we consider even at short $\tau$, $>100$ $^{13}$C nuclei per NV center contribute to the signal. In this regime of a large number of spins, the Bloembergen diffusion model has been shown to excellently capture the spin dynamics [17, 54, 55].

5. **Local and global relaxation**: we have assumed that the $^{13}$C relaxation is set by spatially “local” and “background” sources — by the NV centers and P1 centers respectively. Given that the polarization is injected from the NV center sites, the NVs are assumed to be at the center ($r=0$) of each of the spin systems considered in the ensemble average. On the other hand, the P1 centers are randomly distributed, and in an ensemble average occur with a fixed background density. Moreover, the inter-NV spacing is rather large (25nm), and hence each NV-$^{13}$C combined system can be considered to be unique with minimal overlap (for all $t$ and $\tau$ considered in the paper).

Finally, the values of the various parameters employed in the simulation were set by matching the relaxation data from the experiment.