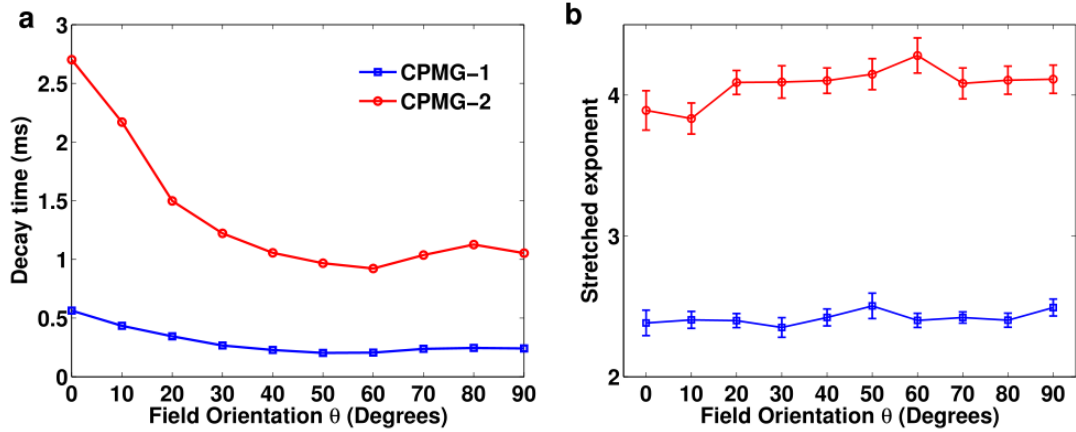


Supplementary Figure 1 | Numerical fits of experimental and theoretical results of $^{nat}\text{Si:P}$ electron spin decoherence by exponential functions $\exp\left[-T/\tau_{\text{ID}} - (T/\tau_{\text{SD}})^2\right]$. (a,c)

Experimental or (b,d) theoretical (solid lines) and fitted (dashed lines) coherence of the P-donor electron spin in the natural ^{29}Si nuclear spin bath under (a,b) CPMG or (c,d) UDD control. Here the same value of $\tau_{\text{ID}} = 10$ ms was used in all the fits. We attribute the deviation seen at ~ 1 ms for CPMG-6 to an overlap with uncorrected stimulated/unwanted echoes. The magnetic field was $B = 0.3$ T applied along the [110] lattice direction.



Supplementary Figure 2 | Theoretical angular dependence of the decay time and stretching factor of central spin decoherence under dynamical decoupling. (a) the decay time τ_{SD} depends strongly on the field orientation. (b) the oscillations of the stretching factor λ with different field orientations are very small compared to the pulse number-parity oscillations. This implies that the pulse number-parity effect can be observed for any field direction. The crystal orientation is done in the [001]-[110] plane with the relations: [001] ($\theta = 0^\circ$), [111] ($\theta = 54.73^\circ$) and [110] ($\theta = 90^\circ$). The error bars correspond to the systematic errors of the stretched exponential fitting of the time-domain spin decoherence.