Supplementary information:

"Electron spin coherence and electron nuclear double resonance of Bi donors in natural Si"

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I. EXPERIMENTAL APPARATUS

Pulsed EPR and ENDOR measurements were performed using apparatus a Bruker Elexsys 680 X-band spectrometer equipped with a low temperature helium-flow cryostat (Oxford CF935). A TWT amplifier was used to provide EPR $\pi/2$ and π pulses of 20 and 40 ns respectively. For ENDOR measurements an Amplifier Research 60 W solid state CW amplifier (0.8–4.2 GHz) and an ENI 100 W (1.5–400 MHz) were used, depending on the frequency range, driven by an Agilent PSG Analogue Signal Generator. RF pulses in the range 4–15 μs were used for a π-pulse on the ^{209}Bi nuclear spin, depending on frequency.

II. SPIN-LATTICE RELAXATION TIMES (T_{1e})

Electron spin-lattice relaxation times T_{1e} were recorded as a function of temperature between 8 and 25 K using a three-pulse inversion recovery sequence $(\pi)_x - \tau - (\pi/2)_{\pm x} - (\pi)_x - (\text{echo})$ and two-step phase cycling. The inversion recovery plots were well fit to a mono-exponential decay and temperature was recorded with a calibrated thermometer below the microwave resonator.

T_{1e} is shown as a function of temperature in Fig. S1, together with the least-squares fit to a model of the form $T_{1e} = A\,T^{-7}$, where $A = 4.3\,(\pm 0.3) \times 10^4\,\text{s}\,\text{K}^7$, consistent with the relaxation being driven by an inelastic Raman phonon scattering process [1]. The T_{1e} data was collected from the $m_\text{I} = -9/2$ line at 577.2 mT; we found T_{1e} to be independent of the choice of m_I.

III. SI:BI ENDOR LINEWIDTHS

Linewidths of the transitions between nuclear sublevels were recorded using a Davies ENDOR sequence: $\pi_\text{mw} - \pi_\text{rf} - \pi/2_\text{mw} - \tau - \pi_\text{mw} - \tau - \text{echo}$. The RF pulse power was optimised by recording nuclear Rabi oscillations while the RF pulse length was chosen to avoid instrumental broadening of the ENDOR lines. ENDOR transition frequencies and linewidths were recorded on each of the ten EPR lines and the resulting distribution of linewidths is displayed in Fig. S2, together with theoretical calculations of the expected line broadening due to (a) strain in the hyperfine coupling to ^{209}Bi and (b) random magnetic fields due to dipolar coupling to ^{29}Si nuclear spins. The observed increase in linewidth at lower
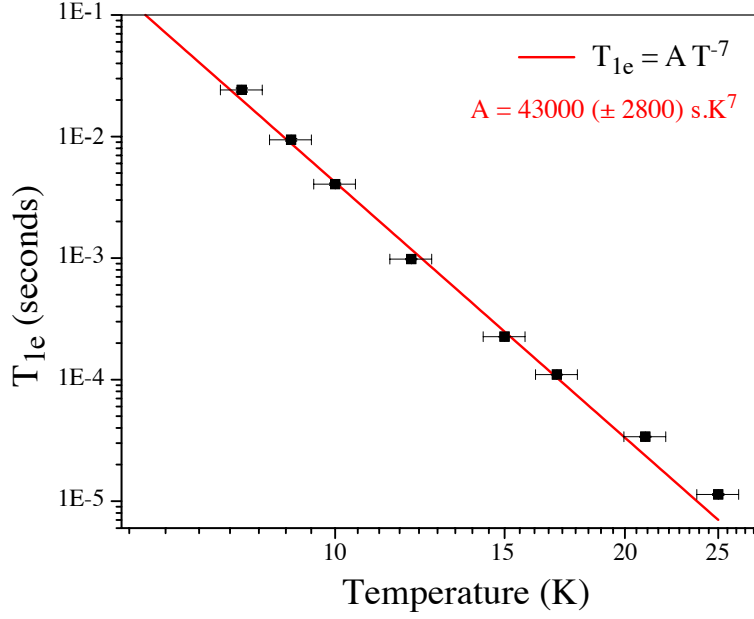
FIG. S1. Temperature dependence of spin-lattice relaxation time in Si:Bi between 8 and 25K, showing a Raman-like T^{-7} behaviour.
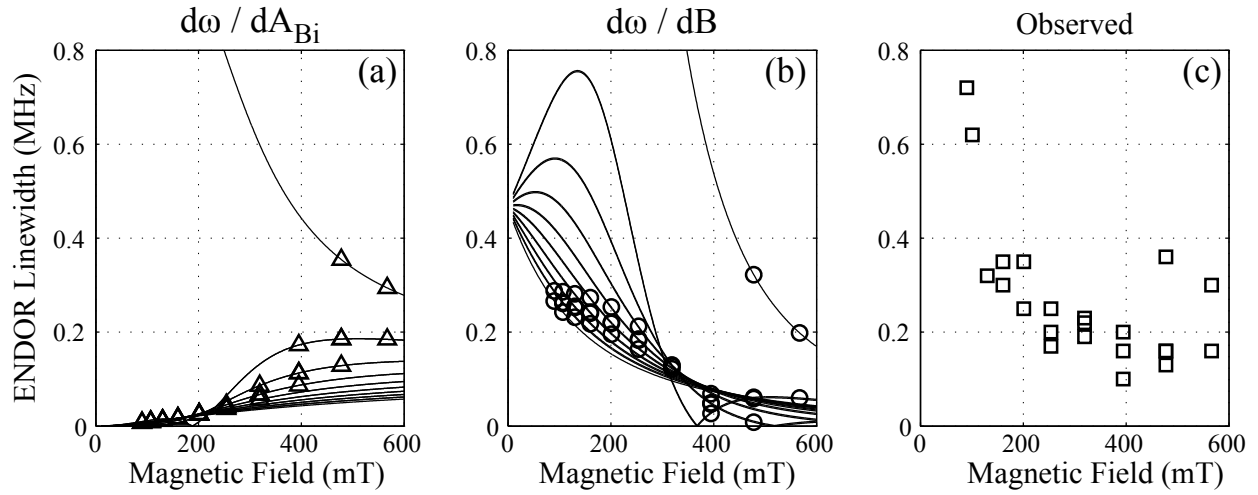

FIG. S2. ENDOR linewidths of hyperfine transitions in the Si:Bi systems observable using a microwave frequency of 9.7 GHz: **(a)** ($\triangle$) predicted due to a 0.23 MHz strain in the ^{209}Bi hyperfine parameter A (corresponding to 0.02%), **(b)** ($\circ$) predicted due to random variation in magnetic field of $\sim$ 2 G. **(c)** ($\square$) Experimental data.
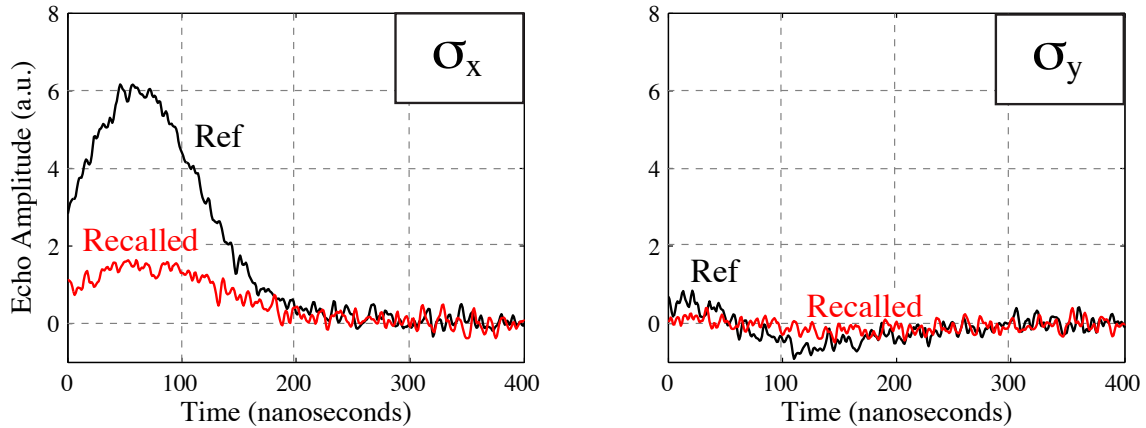
FIG. S3. Electron spin echo transients in Si:Bi comparing a reference (Ref) electron spin coherence with one that is stored in the nuclear spin and then recalled. The loss in signal intensity between the Reference and Recalled echoes is due to the imperfect fidelity of the coherence transfer process. For both cases, the real and imaginary components of the spin echo, measured using a quadrature detector, are shown separately. These signals are proportional to measurements of the σ_x and σ_y components of the density matrix, respectively [3]. Temperature = 14 K

magnetic fields is consistent with the latter mechanism, though A-strain may contribute to ENDOR linewidth at fields above about 400 mT.

IV. FIDELITY OF SI:BI NUCLEAR MEMORY

Using the method described in Ref [2], we demonstrate the ability to store some of the electron spin coherence in the nuclear spin, and subsequently recover it, as shown in Fig. S3. It is not possible to fully excite the ENDOR hyperfine line, leading to a much lower transfer fidelity than observed for P-donors in isotopically purified ^{28}Si, but comparable to that observed for ^{nat}Si:P.

Phase cycling of both microwave and rf pulses is used to confirm the recovered signal is a result of storage in the nuclear spin and is not an artefact such as a stimulated echo.

Comparing the intensity of the recovered electron spin echo with that which was stored allows for a rough assessment of the performance of the transfer — in this case yielding

$I_{\text{stored}} \sim 25\%$.

To compare the transfer fidelity with other quantum memory experiments, we estimate the stored and recovered density matrices and compute the fidelity of that retrieved with respect to that stored using $\mathcal{F} = \text{Tr}\left(\rho_{\text{ref}}\rho_{\text{stored}}\right)$. We neglect the σ_z component in both cases, and use the integrated electron spin echoes in the real and imaginary channels of the quadrature detector to extract σ_x and σ_y components. As in Ref [2], the reference echo is designated as a pure state (i.e. no identity component) and used to normalise the intensity of the recovered echo.

By this method, we estimate the fidelity of the store/retrieve process to be $\mathcal{F} = \left(1 + I_{\text{stored}}\right)/2 = 0.635$. To avoid over-interpreting this result, it should be noted that recovering zero electron coherence (i.e. retrieving only the identity matrix) would yield a fidelity of 0.5.

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