

Non-local synchronization of continuous time crystals in a semiconductor

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Synchronization resulting in unified collective behavior of the individual elements of a system that are weakly coupled to each other has long fascinated scientists. Examples range from the periodic oscillation of coupled pendulum clocks to the rhythmic behavior in biological systems. Here we demonstrate this effect in a solid-state platform: spatially remote, auto-oscillating electron-nuclear spin systems in a semiconductor. When two such oscillators separated by up to 40 μm are optically pumped, their individually different frequencies lock to a common value, revealing long-range coherent coupling. For larger separations, the synchronization breaks. The interaction distance matches the electron spin diffusion length, identifying spin transport as the coupling-mediating mechanism and establishing phase coherence over mesoscopic distances. As a consequence, a wide-area optical pump drives all oscillators within the illuminated spot into a single synchronized state, despite their inhomogeneity. This synchronization accounts for the exceptional stability of the resulting auto-oscillations, enabling collective motion in distributed spin systems and paving the way toward coherent spin networks in spintronics.

INTRODUCTION

Synchronization is a universal phenomenon in which auto-oscillators adjust their rhythms to operate in unison. The defining feature is that auto-oscillators, capable of developing periodic motion, adjust their frequencies through weak interactions while retaining their individuality. Synchronization can arise mutually, when oscillators interact with each other, or be driven by an external periodic force [1, 2]. Huygens' 17th-century observation that two pendulum clocks on the same beam swing in phase demonstrated how mechanical systems can couple through a common medium to lock their motion [3, 4]. Since then, synchronization has been recognized as a general principle across living and non-living systems.

In biology, it underlies fireflies flashing in concert [5], cardiac pacemaker cells beating in harmony [6], and circadian clocks aligning to day-night cycles [7, 8]. In neuroscience, populations of neurons coordinate oscillatory firing to produce brain rhythms [9]. In the physical sciences, synchronization is central to micro- and nano-scale devices: spin-torque nano-oscillators can phase-lock via electrical coupling, enhancing signal power and spectral purity [10–12]. Josephson junctions [13], laser arrays [14], and optomechanical oscillators [15] similarly exhibit mutual entrainment, enabling coherent operation of many units. In atomic physics, synchronization occurs in driven ensembles where global coupling can overcome dephasing. As an example, Wadenpfehl *et al.* [16] showed that thermal rubidium atoms excited to Rydberg states exhibit synchronized oscillations due to mean-field interactions via a shared optical field.

Auto-oscillating systems produce narrow Fourier peaks in the frequency domain, providing an analogy to spatial crystals. So, in crystallography, sharp peaks in momentum space, known as Bragg peaks, arise from constructive interference of waves scattered by atomic

planes, reflecting the underlying periodic lattice structure in space [17]. By this analogy, we refer to an auto-oscillator that exhibits spontaneous, persistent oscillations breaking the continuous time-translation symmetry as a time crystal [18, 19]. A synchronized ensemble of such auto-oscillators can then also be regarded as a time crystal composed of many unit cells. These systems can be divided into discrete time crystals, which are non-autonomous systems responding at subharmonic frequencies under periodic driving [20, 21], and continuous time crystals (CTCs), which are autonomous systems sustained by constant driving and exhibiting self-oscillations. The latter have been realized in many-body systems with long-range interactions, such as Magnon Bose-Einstein condensates [22], Rubidium Bose-Einstein condensates [23], photonic nanomaterials [24, 25], Rydberg gas systems [26], and polariton condensates in semiconductors [27].

Electron-nuclear spin systems in semiconductors can also exhibit time crystal dynamics. Recent work has demonstrated robust auto-oscillations of coupled electron and nuclear spin polarization under continuous circularly polarized excitation, a realization of a continuous time crystal [28]. Furthermore, these oscillations can synchronize to an externally applied modulation when its frequency approaches their natural resonance [29], also showing a subharmonic response. Here, we demonstrate mutual synchronization between autonomous oscillators in a semiconductor system, persisting even when their natural frequencies differ by up to 40%. Distinct spin ensembles (up to 10^9 auto-oscillators) spontaneously lock to a common rhythm, revealing the presence of a robust collective phase. By identifying the underlying mechanism and corroborating it with theoretical modeling, we establish non-local spin synchronization as the genuine collective phenomenon. These results open new pathways toward building coherent spin networks and explor-

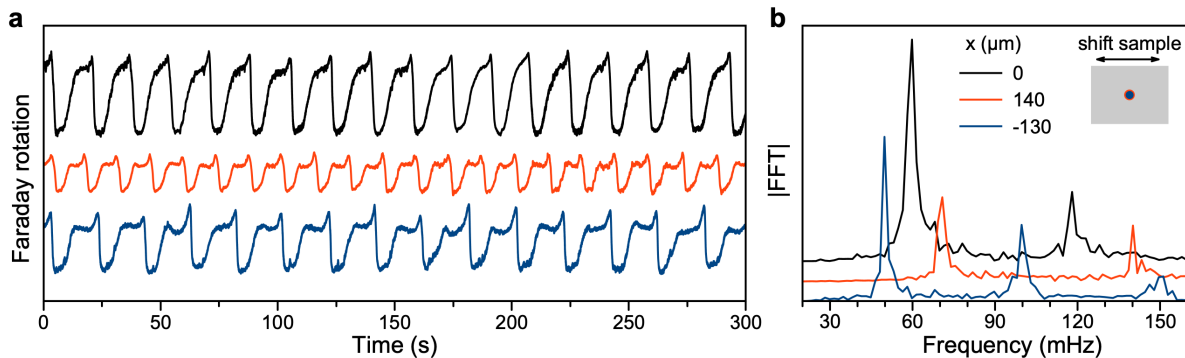


FIG. 1. **Inhomogeneity.** **a**, Examples of time traces measured at different sample positions. Pump and probe spots are overlapped, and the sample is shifted in the range of $-150\ \mu\text{m}$ to $+150\ \mu\text{m}$ along the horizontal direction (x). Pump spot diameter at $1/e^2$ intensity level is equal to $22\ \mu\text{m}$ (FWHM of $13\ \mu\text{m}$). Probe spot size at $1/e^2 = 12\ \mu\text{m}$ (FWHM of $7\ \mu\text{m}$). Pump/Probe powers are 0.15/1 mW, respectively. **b**, Corresponding fast Fourier transform spectra of the traces from panel **a** measured for 10 minutes at the arbitrary position $x = 0$, the top black trace; to the right side of the previous position at $x = 140\ \mu\text{m}$, red trace; and to the left side at $x = -130\ \mu\text{m}$, blue trace. The difference in frequencies is related to the intrinsic sample variation.

ing spatially extended regimes of macroscopic synchronization.

RESULTS

Auto-oscillations

The first step of our study is to establish robust auto-oscillations of the electron-nuclear spin system. Being an open system, the nuclei continuously dissipate energy, yet this loss can be compensated by optical pumping of the donor-bound electron spins [28]. These electrons, oriented by circularly polarized pump light, transfer their spin polarization to the surrounding nuclei through the hyperfine interaction. The resulting nuclear polarization generates an Overhauser field that feeds back on the electron spins. This allows one to monitor the nuclear spin dynamics through the time evolution of the electron spin polarization, measured by the Faraday rotation of the linearly polarized probe laser. For appropriate sample design and experimental conditions, this feedback loop develops non-decaying auto-oscillations of the electron-nuclear spin system (ENSS) - a realization of the CTC [28]. The main experimental parameters and sample features are detailed in the Methods and the Supplementary section 1.

Each donor electron, coupling to $\sim 10^6$ nuclear spins within its wavefunction characterized by the Bohr radius of 11 nm [30], defines the volume of a single auto-oscillator. In effect, the situation is better described by a flux of electrons through the donor that collectively polarize the nuclear spin ensemble, so that the experimentally accessible quantity is the average electron spin polarization at the donor. Finally, the intrinsic variation of the semiconductor sample, related to fluctuations

in donor density and indium incorporation, is expected to result in variations of the observed oscillation frequencies. This feature is a significant advantage of our system due to the systematic variation of the auto-oscillation frequency across the sample.

Figure 1 illustrates such a variation over the sample position. Here, the pump and probe spots are tightly focused (see figure caption) and overlapped, while the sample is shifted in the horizontal direction by up to $\pm 150\ \mu\text{m}$. Figure 1a demonstrates exemplary time traces for three different positions on the sample. The fast Fourier transforms (FFT) for the 10-minute time traces are shown in Fig. 1b, with the measurement positions marked in the panel. The frequency variation is pronounced. To highlight, the number of participating donors in the pump spot focused to a size of $22\ \mu\text{m}$, is on the order of $\mathcal{O}(10^7)$. Despite that, the observed linewidth of the FFT is determined by the inverse measurement time of the non-decaying oscillations, without any dephasing that one could expect for a large number of participating oscillators. This raises the question of whether all the individual auto-oscillators, each defined by a single donor, synchronize and give rise to a macroscopic collective behavior?

Single-pump mutual synchronization

To answer the question whether different auto-oscillations synchronize within a single pump spot, we designed an experiment in which we fixed the sample position and shifted the tightly focused probe spot ($12\ \mu\text{m}$) within the broadened pump ($200\ \mu\text{m}$) for the same range of sample positions as in the previous setting. The pump spot was prepared in a flat-top configuration to provide similar excitation conditions within the excitation spot

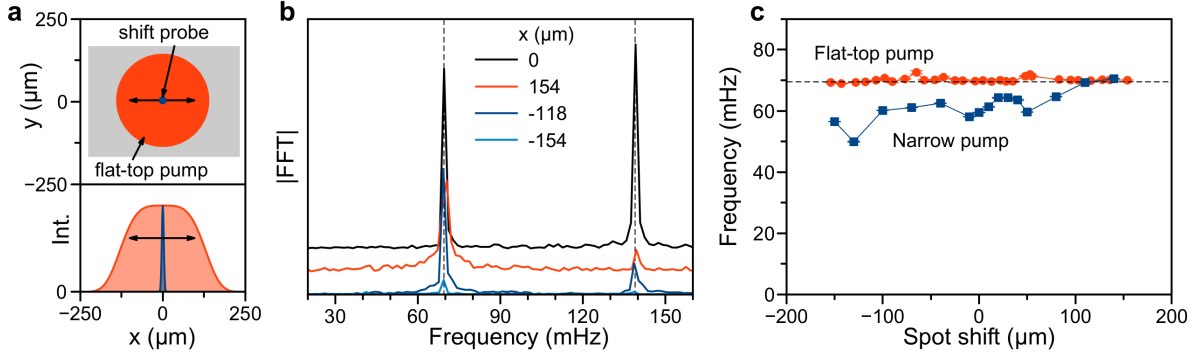


FIG. 2. **Mutual synchronization.** **a**, Sketch of experiment: red pump spot is prepared by a π Shaper to produce a flat-top intensity distribution on the sample, as seen in the simulated bottom part of the figure. Pump spot diameter is about $200\ \mu\text{m}$. The Gaussian probe beam is focused down to $12\ \mu\text{m}$ at $1/e^2$ (FWHM of $7\ \mu\text{m}$) and can be shifted horizontally within the pump beam by the same distances as in the case of Fig. 1a. The bottom part shows the simulated profiles of the lasers. Pump/Probe powers = 10/1 mW. **b**, Examples of the fast Fourier transform spectra of the traces measured for 10 minutes at the center $x = 0$, the top black trace; on the right side of the pump center at $x = 154\ \mu\text{m}$, red trace; and on the left side at $x = -118\ \mu\text{m}$, blue trace, and at $x = -154\ \mu\text{m}$, light blue trace. The difference in frequencies is now negligible compared to Fig. 1b, suggesting complete synchronization within the pump-spot excitation. **c**, Summarized data for the FFT peak positions of the first harmonics for both experimental cases versus shift of the sample for tightly focused pump (blue squares) or the probe shift within a single widened pump (red circles).

diameter, as shown in Fig. 2a. Figure 2b again demonstrates exemplary FFTs for several probe positions inside the pump spot, showing that all observed frequencies across the pump spot are now equal. Figure 2c reflects the difference in the observed FFT frequencies of the first harmonic for both cases: blue squares demonstrate the variation of the frequency over the sample measured with a tightly focused pump spot, while the red circles show that all frequencies are now equalized and are independent of the probe spot position within the single widened pump. These results can be interpreted as a clear demonstration of the mutual synchronization of spatially separated oscillators in an excited ensemble of auto-oscillators. It is essential to note that the relative deviation in frequencies synchronized within a single pump can reach up to 40%. The number of the involved donors in that case can be estimated to be about $\mathcal{O}(10^9)$. Importantly, the present measurements of synchronization within a single pump spot clarify the remarkable robustness of the auto-oscillations reported previously [28]: their stability arises from the emergence of a many-body synchronized state, which protects the dynamics against fluctuations of external parameters. This last demonstration is also well captured by a simplified model in which a single auto-oscillator is used to describe the whole system [28, 29, 31].

Two-pump synchronization

To investigate the processes responsible for the observed synchronization between different auto-oscillators, we modified our experimental setup by adding a second pump laser with the same photon energy as the first one.

Figure 3c demonstrates the setting. We use two tightly focused pump beams (shown by the two small red circles) on the surface of the semiconductor to excite separate auto-oscillations, and a wide defocused probe beam (blue circle) to read out the average electron spin polarization dynamics. The spatial position of one of the pump beams, marked as pump-2, can be precisely shifted on the sample, while pump-1 is always fixed at the center of the probe.

The blue time trace in Fig. 3a demonstrates several minutes of the auto-oscillations at the pump-1 position (pump-2 is blocked), where non-decaying oscillations are observed. The corresponding FFT for a ten-minute time trace is then represented in Fig. 3b by the blue curve, showing the first harmonic peak at $f_1 = 91\ \text{mHz}$. The red colored time trace in the middle of Fig. 3a demonstrates a pump-2-only experiment, where pump-2 is, as an example, shifted by $-25\ \mu\text{m}$ away from the pump-1 position, and pump-1 is blocked. The signal intensity is shown on the same scale. It is smaller than that of the top trace of pump-1, which, in addition to the variation of the parameters of the sample, is also related to a weaker probe intensity at this spatial position, see the intensity profile variation at the bottom part of Fig. 3c. Figure 3b shows the corresponding FFT with $f_2 = 69\ \text{mHz}$, 22 mHz lower than at the pump-1 position, by the red-colored spectrum. Finally, the green-colored trace at the bottom of Fig. 3a reflects the case when both pump-1 and pump-2 are open simultaneously. As shown in Fig. 3b, the corresponding FFT exhibits a single peak at $f_{\text{sync}} = 80\ \text{mHz}$, located between f_1 and f_2 , demonstrating clear synchronization. In that case, the FFT does not necessarily have to show the average frequency, but there is clearly only one peak in the 1st-harmonic range between f_1 and f_2 .

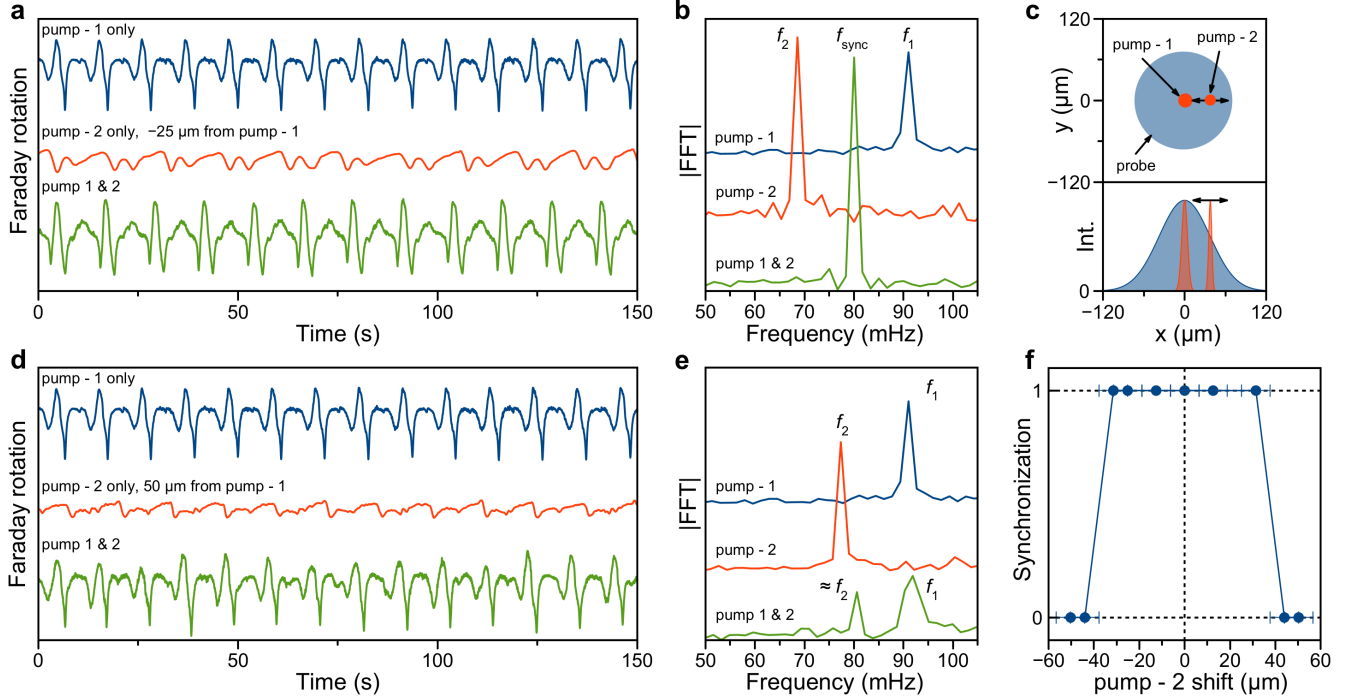


FIG. 3. Range of synchronization. **a**, Experimental time traces of auto-oscillations for three different cases: the top blue one shows the pump-1 only, the red one in the middle is the pump-2 only, shifted horizontally by $-25 \mu\text{m}$ away from the pump-1 position. Finally, the last green time trace at the bottom represents the combined application of pump-1 and pump-2. See Fig. 3c for the labeling of the pumps. **b**, First harmonic range of the fast Fourier transform spectrum of the traces presented in panel **a**, measured for 10 minutes. As one can see, each separate pump beam at its sample position induces auto-oscillations with a characteristic frequency. If both pumps are applied together, one can see a single frequency, a hallmark of synchronization. Laser powers for Pump-1/Pump-2/Pr = 0.1/0.1/1 mW. **c**, Sketch of three Gaussian laser spots on the sample with the wide blue spot representing the probe ($1/e^2 = 150 \mu\text{m}$, FWHM of $88.3 \mu\text{m}$), the central red spot is the pump-1 ($1/e^2 = 17 \mu\text{m}$, FWHM of $10 \mu\text{m}$), and the red spot on the right is the pump-2 ($1/e^2 = 9 \mu\text{m}$, FWHM of $5.3 \mu\text{m}$) that can be shifted horizontally. The top sketch shows a frontal view of the sample, and the bottom sketch shows the normalized intensity profile simulation of the involved beams of $38 \mu\text{m}$ separation. **d**, Experimental time traces of auto-oscillations for the case of the pump-2 spot shifted horizontally by $50 \mu\text{m}$ away from the pump-1 position. **e**, First harmonic range of the fast Fourier transform spectrum of the traces presented in panel **d**, measured for 10 minutes. Each separate pump beam induces auto-oscillations with different frequencies. When both pumps are applied together (green, bottom), two frequencies are observed at positions close to those of the single-pump beam cases, indicating no averaging and thus no synchronization. **f**, The measured synchronization range presented for a pump-2 shift from $-50 \mu\text{m}$ to $+50 \mu\text{m}$ relative to pump-1 position. The synchronization radius is about $38 \pm 3 \mu\text{m}$ measured between the pump-spot centers. The precision of the rotation screw determines the error bars.

We emphasize that the green-colored time trace in Fig. 3a represents a novel type of oscillation that cannot be composed of a combination of two contributing signals. See the Supplementary section 3 for a wider frequency range than in Fig. 3a.

Range of synchronization

We now continue shifting the pump beams apart. Figure 3d illustrates the case where the pump-1-only signal is presented again, but the pump-2-only signal is shifted $50 \mu\text{m}$ away from the pump-1 position (see the red-colored time trace). The green-colored bottom trace shows the signal with both pumps applied together. Figure 3e shows the corresponding FFT spectra with a main

difference relative to Fig. 3b: the both-pumps-on case demonstrates two frequencies close to those of the positions of the contributing signals, indicating the absence of synchronization. In that example, one can argue that the green-colored trace in Fig. 3d represents the superposition of both measured signals.

Finally, Fig. 3f summarizes all measurements with the pump-2 position scanned across the range of $-50 \mu\text{m}$ to $+50 \mu\text{m}$ of relative shift between the pump spots. Here, the synchronization is present (1) if one observes a single frequency for the case of both pumps being applied together, and the synchronization is absent (0) if one detects both contributing single-pump frequencies under illumination with two pumps. We conclude that the synchronization range is $38 \pm 3 \mu\text{m}$ taken between the central positions of the pump spots, or taking the spot sizes

of the pump beams into account (given in the caption of Fig. 3), the beam separation is $25 \pm 3 \mu\text{m}$ defined as the distance between the points where the spot intensities fall to $1/e^2$ of their maximum value. In Supplementary section 3, we additionally demonstrate the appearance of intermodulation products in the superposition signal without synchronization and the FFT spectra for all pump-to-pump distances shown in Fig. 3f.

ORIGIN OF SYNCHRONIZATION

For two spatially separated, auto-oscillating ENSSs, we only need a weak interaction that makes the instantaneous frequency (or phase) of one depend on the other. Let's consider the most likely mechanisms present in an n -doped GaAs semiconductor that could provide a connection between remote places:

- **Nuclear spin diffusion:** Slow flip-flop-driven transport of nuclear polarization spreads Overhauser fields between the spots. The diffusion constant for GaAs is tiny ($D_N \sim 10^{-13} \text{ cm}^2/\text{s}$) [32] and is in the same range as the one determined for our sample [30], so the coupling is slow (seconds-minutes) but long-lived. Using the given D_N and the longest measured value of the nuclear spin relaxation time for our sample $\tau_N = 210 \text{ s}$ [30], leads to a diffusion length of $L_N = \sqrt{D_N \tau_N} \approx 46 \text{ nm}$.
- **Electron mediated coupling:** Hopping electrons can correlate nuclear baths in separated regions, altering nuclear spin polarization and effectively coupling the ENSS phases. For a given concentration of about 10^{16} cm^{-3} the diffusion coefficient is estimated to be about $D_{\text{hop}} \approx 1 \text{ cm}^2/\text{s}$ [33], which taken with the spin relaxation time of $\tau_s = 120 \text{ ns}$ leads to the length of $L_{\text{hop}} \approx 3.5 \mu\text{m}$.
- **Electron spin diffusion:** Spin-polarized free carriers created in one spot diffuse into the other, affecting the donor-bound electrons by the spin-exchange coupling [34]; the resulting Overhauser field feedback pulls the precession frequency and can lock the two oscillators. The spin-diffusion length in this case is given by $L_s = \sqrt{D_s \tau_s}$, where D_s is the spin diffusion constant and τ_s is the electron spin relaxation time. Taking into account the known values of $D_s = 24 \text{ cm}^2/\text{s}$ [35] and $\tau_s = 120 \text{ ns}$ measured for our sample [30], one arrives at the diffusion length $L_s \approx 17 \mu\text{m}$.

To summarize, based on the experimentally observed synchronization range of $\sim 40 \mu\text{m}$, we can suggest that the primary mechanism responsible for the synchronization is the spin diffusion of free electrons [33, 34]. See also Supplementary section 4 for another excluded mechanism.

To simulate it, we extend the model presented in the Refs. [28, 31, 36]. The Bloch equation describing the spin

polarization ($\mathbf{S}$) precession in the external field (B_{ext}) and the Overhauser field (B_N) is extended by a term accounting for spin diffusion [37]:

$$\mathbf{S} = \mathbf{S}_0(x) + \frac{g\mu_B T_s}{\hbar} (\mathbf{B}_{\text{ext}} + \mathbf{B}_N) \times \mathbf{S} + D_s T_s \frac{\partial^2 \mathbf{S}}{\partial x^2}. \quad (1)$$

Here, D_s is the spin diffusion coefficient, $\mathbf{S}_0(x)$ is the average electron spin polarization in the absence of the magnetic field, created by the continuous wave pump with Gaussian or flat-top spatial profile, μ_B is the Bohr magneton, and g is the electron g -factor. $\hbar/g\mu_B T_s$ is the half-width at half-maximum of the electron Hanle curve that is not influenced by dynamic nuclear polarization. T_s is the electron spin relaxation time, $\hbar$ is the reduced Planck constant. For clarity, we restrict the diffusion to a single spatial direction (x).

The precession of the electron spin polarization about the total magnetic field changes the Overhauser field in time according to [36, 38]:

$$\frac{d\mathbf{B}_N}{dt} = -\frac{1}{T_N} (\mathbf{B}_N - \hat{a}\mathbf{S}), \quad (2)$$

where $\hat{a}$ is the second-rank tensor describing the process of dynamic nuclear polarization. The model suggests that $\hat{a}\mathbf{S}$ is a linear function of $\mathbf{S}$. The tensor has been simplified in the lowest order approximation (for details, see Methods section, Ref. [28], and the corresponding Supplementary section 5).

To account for sample variation, we introduce a spatial distribution of the hyperfine interaction parameters (see Supplementary section 5). When the system is excited by a spatially narrow Gaussian pump, auto-oscillations emerge, as shown in blue in Fig. 4a, with their frequency extracted via FFT (blue curve in Fig. 4b). Excitation by pump-2, positioned $0.5L_s$ away from pump-1, produces a modified oscillation pattern due to the inhomogeneous hyperfine coupling, with the corresponding frequency shift illustrated in red in Fig. 4b (L_s is the spin diffusion length.) When both pump-1 and pump-2 are applied simultaneously, the auto-oscillations occur at an intermediate frequency between f_1 and f_2 , as seen in Fig. 4b (green). This behavior represents a clear manifestation of synchronization of auto-oscillations mediated by spin diffusion.

Synchronization mediated by spin diffusion is sensitive to the spatial separation between pump-1 and pump-2. To illustrate this, pump-2 was placed $2L_s$ away from the position of pump-1. The auto-oscillations induced by pump-2 are shown in red in Fig. 4c, with their corresponding frequency indicated in red in Fig. 4d. When both pumps are applied simultaneously, the FFT exhibits two distinct peaks close to f_1 and f_2 , as shown in Fig. 4d, with the corresponding time trace also presented in Fig. 4c. The emergence of two distinct frequencies under simultaneous excitation by pump-1 and pump-2 is a clear indication of the loss of synchronization. Figure 4e shows the synchronization range versus

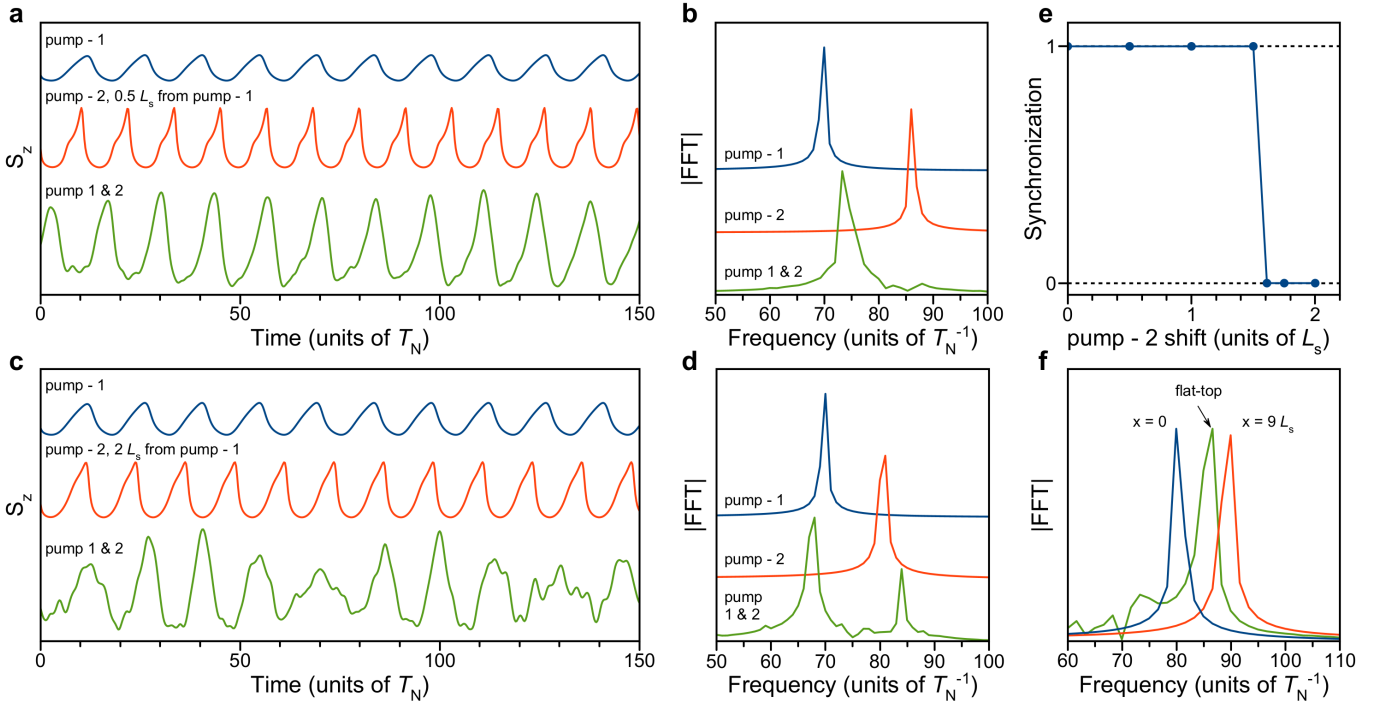


FIG. 4. **Simulation of synchronization.** **a**, Simulated time traces of auto-oscillations for three cases. The top (blue) trace corresponds to pump-1 only, the middle (red) trace to pump-2 only, shifted by $0.5L_s$ from the pump-1 position. The spatial width (FWHM) of both pump-1 and pump-2 is $0.25L_s$. The bottom (green) trace shows the combined excitation by pump-1 and pump-2. See Fig. 3c for pump labeling. Detection is performed with a spatially wide probe covering the range $0 - 10L_s$. **b**, First harmonic of the fast Fourier transform spectra of the traces in panel **a**. **c**, Simulated time traces of auto-oscillations for three cases: the top (blue) trace corresponds to pump-1 only, the middle (red) trace to pump-2 only, shifted by $2L_s$ from the pump-1 position, and the bottom (green) trace to their combined excitation. **d**, First harmonic of the FFT spectra of the traces in panel **c**. **e**, Extracted synchronization range for pump-2 shifts between 0 and $2L_s$ relative to pump-1. **f**, Synchronization in simulation under flat-top pump excitation. The blue curve shows the first harmonic in the FFT for the spatially narrow pump-1 and narrow probe, the red curve corresponds to pump-2 and probe shifted by $9L_s$, and the green curve shows the resulting signal under flat-top pump excitation spanning $0 - 10L_s$. The detection is performed with a spatially narrow probe at $9L_s$.

pump-2 shift. Synchronization persists up to $1.5L_s$, consistent with experiment (Fig. 3f). The simulations reveal that the difference between the maximum and minimum values of the hyperfine coupling parameter significantly affects the synchronization threshold (see Supplementary section 5). So, for more similar initial frequencies, the synchronization range expands.

For completeness, we also simulated synchronization under a flat-top pump covering the spatial range from $x = 0$ to $x = 10L_s$. To illustrate the spatial variation of the hyperfine interaction, we show the auto-oscillation frequencies f_1 and f_2 obtained for a spatially narrow pump placed at $x = 0$ and $x = 9L_s$, respectively (blue and red curves in Fig. 4f). Under flat-top excitation, the auto-oscillation frequency locks to an intermediate value between f_1 and f_2 across the entire range $0 \leq x \leq 10L_s$ (see Fig. 4f). Flat-top excitation demonstrates that spin diffusion promotes global synchronization across an inhomogeneous sample, consistent with the experimental observations.

Together with the agreement between experiment and simulation, the results of simulations establish electron spin diffusion as the microscopic mechanism underlying the collective synchronization of auto-oscillations. However, to obtain a more complete picture, we also determined the diffusion length using the independent method of time-resolved magneto-optical Kerr rotation microscopy (see the Method section and the Supplementary section 6). The obtained spin diffusion value of $D_s = 28 \pm 1 \text{ cm}^2/\text{s}$ is very close to the previously reported one $D_s = 24 \text{ cm}^2/\text{s}$ [35] cited above. This corresponds to a diffusion length of $L_s = 18.3 \pm 0.3 \mu\text{m}$ in our sample, confirming the observations.

CONCLUSION

Our results establish that spatially separated electron-nuclear spin systems in a semiconductor can mutually synchronize through electron spin diffusion, locking

their oscillations over mesoscopic distances. This collective coherence persists under wide-area excitation, where an extended ensemble of auto-oscillators contributes to a single synchronized state despite the environmental variation. Importantly, the emergence of such macroscopic synchronization is itself the key factor that stabilizes the auto-oscillations, protecting them against fluctuations and disorder that would otherwise disrupt coherent dynamics. Demonstrating that diffusive spin currents can mediate both long-range phase coherence and enhanced stability highlights a new route for engineering

robust networks of coupled spin oscillators in solid-state platforms. Furthermore, integrating electrostatic gating or lateral drift control would convert this passive non-local link into an actively tunable coupling channel. Such controllable networks may serve as a basis for analog neuromorphic architectures, where information is processed through collective phase dynamics rather than discrete logic states. Beyond deepening our understanding of non-equilibrium collective dynamics, spin-mediated synchronization offers prospects for coherent spintronic architectures and on-chip time-crystal-based devices.

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METHODS

Setup

Ref. [28] and the corresponding Supplementary sections 1 and 2 describe our sample and experimental setup in all details. Here, we reiterate the most relevant and deviating parts. We use the continuous wave laser diode emitting at 1.579 eV (785 nm) for the pump laser, which is then routed through a quarter-wave plate to create circular polarization. The linearly polarized probe laser is produced by the continuous wave Ti:Sapphire ring-laser and is fixed at the 1.426 eV (869.4 nm). Both lasers are combined on a non-polarizing beam splitter and are focused by a single lens at the sample. Depending on the experiment, we also split a portion of the pump laser to produce an additional pump beam, which hits the sample at an angle of approximately 30 degrees. For the two-pump version presented in Fig. 3, we use an additional beam as a pump-1, which is fixed at the center of the probe beam at the sample. The second part of the pump beam, which is routed through the rotatable NPBS and combined with the probe, is then referred to as a pump-2. The NPBS controls the pump-2 position at the sample by the tip-tilt-rotation stage (TTR001 Thorlabs). For the experiment, presented in Fig. 1, we use only one pump beam and shift the whole cryostat by the micrometer stages. For the experiment in Fig. 2, the pump-1 beam is going through the π Shaper (AdlOptica Focal- π Shaper), which converts the Gaussian beam to a flat-top profile. The position of that beam is fixed at the sample, while the probe position is shifted within the pump.

The sample is mounted in a helium flow cryostat at $T = 6$ K in the center of two orthogonal pairs of electromagnet coils, generating the magnetic field components $B_x = -1$ mT and $B_z = 0.176$ mT.

The GaAs substrate of the sample completely absorbs the pump laser, while the probe laser is transmitted and then analyzed by the polarization bridge, which is built with a half-wave plate and a Wollaston prism. A balanced photodiode is used to measure the Faraday rotation of the plane of the linearly polarized probe.

We have used the Beam-master scanning multiple knife-edge device (Coherent) to characterize the beam's spot sizes at the sample position. It allows for measuring beam spot diameters as small as $3 \mu\text{m}$ with $0.1 \mu\text{m}$ resolution and larger beams up to 3 mm with $1 \mu\text{m}$ resolution.

More details and schematics are provided in the Supplementary section 2.

TR-MOKE

For the spatial mapping and extraction of the spin diffusion coefficient, we use the following experimental configuration. Initial laser pulses with a temporal width

of ~ 35 fs are derived from a 60-MHz mode-locked Ti:sapphire oscillator. They are split into pump and probe paths and tuned by grating-based pulse shapers, resulting in pulses with a bandwidth of ~ 0.5 nm and temporal resolution of ~ 1 ps. Pump pulses are modulated between left and right circular polarizations by an electro-optic modulator. The linearly polarized probe pulses are collinear with the pump and focused on the sample surface through a microscope objective. The FWHM diameters of pump and probe pulses are 3 ± 0.1 and 1 ± 0.1 μm , respectively. Kerr rotation is measured using balanced photodiodes connected to a lock-in amplifier, referenced to the modulation frequency. The probe polarization is resolved using a half-wave plate and a Wollaston prism. A mechanical delay stage adjusts the delay time between the pump and probe. The spatial overlap of the pump with the fixed and centered probe is adjusted through a lateral translation of the input lens of the beam-expanding telescope in the pump path [39, 40]. All measurements are performed with the pump photon energy set to $E_{\text{pu}} = 1.459$ eV and the peak power density of 4.7 MW/cm². The probe photon energy is $E_{\text{pr}} = 1.43$ eV with peak power density of 1.2 MW/cm².

Details on simulation model

The quadrupole unperturbed nuclear spin sublevels create the Overhauser field $\mathbf{B}_N^0$ as in pure, unstressed GaAs. $\mathbf{B}_N^0$ is aligned along the external magnetic field and compensates for the Zeeman splitting of the electrons in the external magnetic field. Due to the strong deformation caused by the indium incorporation, the spin of the i -th nucleus is oriented along the main local axis $\mathbf{n}_i$ of the tensor describing the quadrupole interaction rather than along the external magnetic field. The contribution of these nuclei to the total Overhauser field is $\mathbf{B}_Q = \sum_i a_i (\mathbf{S} \mathbf{n}_i) \mathbf{n}_i$, where the summation is carried out over all quadrupole perturbed nuclei within the electron localization volume around a donor. For an isotropic distribution of the axes, the field can be written as $\mathbf{B}_Q = a_N \mathbf{S}$. Therefore, $\hat{a}$ can be reduced to the simplified form: $\hat{a} \mathbf{S} = \mathbf{B}_Q + \mathbf{B}_N^0 = a_N \mathbf{S} + b_N (\mathbf{S} \mathbf{h}) \mathbf{h}$, where b_N is the parameter of the hyperfine interaction between the electrons and the nuclei, and $\mathbf{h}$ is the unit vector of the externally applied magnetic field. The tensor components are: $\hat{\alpha}_{\alpha\beta} = a_N \delta_{\alpha\beta} + b_N h_\alpha h_\beta$, with $\alpha, \beta = x, y, z$ coordinates.

The solution is integrated over a time window of $\sim 3T_s$, since the nuclear spin system has the relaxation time of $T_N \gg T_s$ and therefore experiences a stationary electron-nuclear polarization. This approach avoids numerical difficulties associated with solving the stationary Bloch equation directly.

The simulation algorithm consisted of three steps. In the first step, the electron spin polarization and its diffusion were calculated in the presence of the external magnetic field only. The stationary Bloch equation given

by Eq. (1) leads to a divergent solution. To eliminate this divergence, the time-dependent Bloch equation was solved, and the resulting steady-state value was obtained by averaging over a time interval of $3T_s$. This procedure is justified because the nuclear spin relaxation time is much longer than the electron spin lifetime. The resulting spatial distribution of electron polarization defined the spatial profile of the Overhauser field, which exhibited auto-oscillations according to Eq. (2). In the final step, the diffusion of the electron spin polarization was computed in the total magnetic field (Eq. (1)), given by the sum of the external field and the Overhauser field obtained in the previous step.

All calculations were performed numerically in Matlab using standard built-in libraries. The Overhauser field was calculated using the ode23 solver, while spin diffusion was simulated with the pdepe solver on a 600×4000 grid (space $\times$ time), covering a spatial range of $11.5L_s$ and a temporal range of $1000T_N$ with Neumann boundary conditions. The resulting signal included a transient region corresponding to the stabilization of auto-oscillations, containing approximately 250 points in the time series. The Fourier spectra presented in the main text were calculated after removing these transient points.

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AUTHOR CONTRIBUTIONS

A.G. and N.E.K. contributed equally to this paper. A.G. built the experimental apparatus and performed the measurements together with Ph.A.H. and L.K. B.W.G. and S.A. provided the measurements and analysis of the scanning Kerr microscopy. N.E.K. and A.G. analyzed the data. N.E.K. and V.L.K. provided the theoretical description. A.G., N.E.K., and V.L.K. wrote the manuscript in close consultation with M. Betz and M. Bayer.

COMPETING INTERESTS

The authors declare no competing interests.

Supplemental Materials for Mutual synchronization of continuous time crystals in a semiconductor

S1. SAMPLE CHARACTERIZATION

Figure S1 shows the normalized photoluminescence (PL) of the used sample. In comparison to previous publications [S28, S29], we have used the same sample but etched a majority of the GaAs substrate away to test its effect on the behavior of the auto-oscillations. It has caused a shift in the PL emission to a higher wavelength. The dashed red line demonstrates the position of the PL in its initial form, while the blue solid line shows the position after the etching process. It had no observable effect on the doping and overall CTC stability and operation, except that the main frequency of the CTC (and corresponding higher harmonics) has been decreased, from ~ 160 mHz to ~ 90 mHz.

S2. EXPERIMENTAL SETUPS

The experimental setup for a two-pump configuration, presented in Fig. 3 of the main text, is shown in Fig. S2a.

We use the continuous wave laser diode emitting at 1.579 eV (785 nm) for the pump laser, which is then routed through the laser power stabilizer (NEL03A Thorlabs). We split the pump beam into pump-1 and pump-2 using NPBS. For the pump-1 beam, we use a lens with $f = 35$ mm, a $20\ \mu\text{m}$ pinhole, and collimate the beam with a 150 mm lens. After that, we have beam routing to $\lambda/2$ -Glan-Taylor (GT)- $\lambda/4$ with wide apertures (15 mm) and focus it at the sample using an achromatic lens with a focal length of $f = 100$ mm. The beam is directed at an angle of approximately 30° and is focused to a diameter of $17\ \mu\text{m}$ at a $1/e^2$ intensity roll-off. The FWHM is the diameter at $1/e^2$ times $\sqrt{\ln 2}/\sqrt{2}$.

The pump-2 is prepared by using 35 mm lens, then $20\ \mu\text{m}$ pinhole, and 150 mm collimating lens. It is then going through GT- $\lambda/4$ with wide apertures (pupil diameter 15 mm) to the NPBS, where it is combined with the probe beam path.

The linearly polarized probe laser is created by the continuous wave Ti:Sapphire ring-laser and is fixed at the energy of 1.426 eV (869.4 nm).

The pump-2 and probe beams are focused by a single achromatic lens of 60 mm focal length at the sample.

The $10\ \mu\text{m}$ epilayer and the remaining GaAs substrate of the sample absorb entirely the pump laser. In contrast, the probe laser is transmitted and then analyzed by the polarization bridge, which is built with a half-wave plate and a Wollaston prism. A balanced photodiode is used to measure the Faraday rotation of the plane of the linearly polarized probe, see Fig. S2a. The differential signal is digitized with 4 ms time steps and stored on the PC. All

presented time series in the main text were smoothed using a 20-point moving average.

For the measurements of sample inhomogeneity (see Fig. 1), we block the pump-1 beam (keep the pump-2 beam on) and shift the cryostat using the μm stages.

For the flat-top pump experiment (see Fig. 2), we use the scheme shown in Fig. S2b. Here, the pump-2 beam is blocked, while the probe is routed in the side entrance of the NPBS to be shifted at the sample. Pump-1 is transformed using the π Shaper to produce the flat-top beam at the sample.

To recalculate the shift of the pump by rotating the stage with NPBS in Fig. S2a, one has to consider the following: a shift at the sample $x = f \cdot 2 \cdot \tan(\theta)$, and for small θ , $\tan(\theta) \approx \theta$. f - is the focal length of the lens, in our case 60 mm. One full rotation of the μm -screw at the rotation stage corresponds to $500\ \mu\text{m}$ scale reading and 1.5° of rotation about vertical axis, or $1\ \mu\text{m} = 3 \times 10^{-3} \text{ deg}$. Therefore, a shift at the sample $x = f \cdot 3 \times 10^{-3} \cdot 2\pi/180 \cdot s[\mu\text{m}]$, with s - being the micrometer scale of the rotation screw. With $f = 60$ mm it is: $x = 2\pi \cdot s[\mu\text{m}]$. The scaling factor was additionally confirmed by placing the μm scale ruler at the sample position and reading the spot shift using the camera.

As the probe beam also passes through the same (rotating) NPBS, it is expected to exhibit a weak horizontal shift with NPBS rotation, which is an order of magnitude smaller than that of the reflected pump beam and is therefore irrelevant in all experiments.

S3. EXTENDED DATA

Depending on the symmetry and strength of coupling, two auto-oscillators can produce the following response in the FFT:

- **1:1 synchronization:** a single, narrow line at f_{sync} ; harmonics at integer multiples of f_{sync} ; intermodulation sidebands largely vanish, see Fig. S3a. The synchronization frequency between two auto-oscillators is determined by the balance of coupling strengths [S41], nonlinear frequency shifts, dissipation, and nonreciprocal feedback [S42].

$$f_{\text{sync}} \approx f_1 + \frac{\kappa_1}{\kappa_1 + \kappa_2}(f_2 - f_1), \quad (\text{S3})$$

where κ_i are effective coupling weights. Thus, the synchronized frequency interpolates between the two natural frequencies, leaning toward the one with stronger coupling, higher amplitude, or lower damping.

- **Frequency pulling:** the two peaks move toward each other and broaden. Partially seen in see Fig. S3b for the fundamental frequencies and higher harmonics.
- **Free-running:** two distinct peaks at f_1 and f_2 ; in a combined signal you may also see intermodulation products ($nf_1 \pm mf_2$) and a time-domain beat at $(|f_1 - f_2|)$, see Fig. S3c, Fig. S4, and Fig. S5a.

Figure S3a shows the extended frequency range, demonstrating how the synchronization is seen for the first three harmonics of the signal. The green colored trace demonstrates peaks in the middle between the pump-1-only and pump-2-only harmonics.

Figure S3b shows the extended frequency range in the case where the synchronization is broken. The green-colored trace demonstrates a double-peaked structure for each harmonic, with one peak corresponding to the frequency of pump-1 and the second peak being close to the pump-2-only position (although slightly shifted).

Figure S3c is the expanded view of Fig. S3b for the case without synchronization with vertical lines marking the main harmonics, the beat, and intermodulation products.

Figure S4 demonstrates the FFT peaks positions for all cases of two-pump beam separations, presented in Fig. 3f of the main text. The presence of a single peak in the both-pumps-on case indicates synchronization, although the peak position does not necessarily coincide with the exact average of the individual frequencies. In the absence of synchronization, two distinct peaks corresponding to the contributing frequencies, along with their intermodulations, can be observed.

S4. INTERACTION BY CROSS-ILLUMINATION

A pump light scattered from one spot could reach the other spot and provide a (very) weak common drive. This regime cannot be distinguished from the others in an experiment with a single pump, as shown in Fig. 2, where different ENSSs are excited using a single pump spot. Still, we can be certain of a clear separation between the spots in experiments with two pumps, as shown in Fig. 3, excluding this mechanism.

To demonstrate the influence of the cross-illumination, we have expanded both pump spots as shown in Fig. S5c. Figure S5a demonstrates exemplary FFT spectra at different pump-separations, while Fig. S5b summarizes the measurements. In that setting, the observed synchronization radius between the pump spot centers has increased to about $50 \pm 5 \mu\text{m}$. Taking the spot sizes into account, this leads to a beam separation of $19 \pm 5 \mu\text{m}$, defined as the distance between the points where the spot intensities fall to $1/e^2$ of their maximum values. This value is close to that observed for the spatially narrow pump spots of $25 \pm 3 \mu\text{m}$, and gives additional support for our observations.

S5. SPIN DIFFUSION CALCULATIONS

Figure S6a shows the spatial profile of the flat-top pump covering the region from $x = 0$ to $x = 10L_s$ used for calculations of Fig. 4f in main text. The corresponding components of the electron spin polarization in the external magnetic field $B_x = -1 \text{ mT}$ tilted by $\alpha = 10^\circ$ with respect to the sample plane, S_x (black), S_y (blue), and S_z (red), are shown in Fig. S6b.

For calculations of Figs. 4a-e, the excitation of two Gaussian pumps with a full width at half maximum of $0.25L_s$, separated by a variable distance, is used. Figure S6c illustrates their spatial profile. The components of the electron spin polarization, S_x (black), S_y (blue), and S_z (red), are shown in Fig. S6d for excitation by the two Gaussian pumps in the external magnetic field $B_x = -1 \text{ mT}$ tilted by $\alpha = 10^\circ$ with respect to the sample plane.

Figure S7a shows the spatial dependence of a_N (red) and b_N (blue) under flat-top pump excitation (green), used for calculations of Fig. 4f in the main text. Figure S7b presents the corresponding spatial dependence of the first harmonic calculated for the hyperfine interaction parameters shown in panel a in the absence of spin diffusion in the external magnetic field $B_x = -1 \text{ mT}$ tilted by $\alpha = 10^\circ$ with respect to the sample plane. Figure S7c displays the spatial dependence of a_N (red) and b_N (blue) under excitation by two Gaussian pumps separated by a distance of $2L_s$, with the green curve indicating the respective pump profiles. The relative values of the hyperfine interaction parameters were increased compared to those used in the calculations with the flat-top pump. This adjustment was necessary because the spin polarization generated by a spatially narrow Gaussian pump is smaller than the average polarization produced by the spatially broad flat-top pump. To ensure that the resulting frequency of the first harmonic in the Fourier spectrum remains comparable between the two calculation types, the hyperfine coupling constants were adjusted.

To highlight the dependence of the synchronization range on the hyperfine coupling span, we present an additional calculation. Figure S8 shows an example of the spatial distribution of hyperfine interaction parameters. The difference between the minimum and maximum values of a_N is now increased up to 50 mT , which is larger than in the distribution used for the calculations presented in the main text. In this case, the synchronization threshold shifts down to $0.65 L_s$.

S6. SPATIAL KERR MICROSCOPY

Figures S9a and S9b demonstrate the spatial maps of the time-evolution of the excited spin polarization measured by the pump-probe scanning Kerr microscopy at time delays of 0.2 and 1.8 ns , exemplary. The images directly reveal the spatial dependence of spin diffusion after

the excitation pulse of 1 ps duration.

Figure S9c demonstrates the exemplary space-resolved spin polarization, derived from the measured Kerr rotation at 0.2 ns time delay in the spatial Kerr microscopy. As shown, the space-resolved polarization consists of two Gaussian profiles: a narrow one is produced by the pump pulse at zero time delay and becomes broader with increasing time delay. In contrast, the wider profile is related to the signal coming from the previous-pump-pulse contributions, similar to Ref. [S43]. We have fitted both of them separately. The corresponding dependencies of the FWHM² versus time delay are shown in Figs. S9d and S9e, yielding similar values of diffusion coefficients: $D_s = 28 \pm 1 \text{ cm}^2/\text{s}$ and $D_s = 35 \pm 6 \text{ cm}^2/\text{s}$, respectively.

Furthermore, following the discussion in Ref. [S43], one

can estimate the influence of hot electrons. As excitation occurs at much higher energies than the detection, the pump excites hot electrons that relax/cool down with an accelerated diffusion constant. According to Ref. [S43], one expects the electrons to cool down in approximately 400 ps, while the diffusion constant is changing exponentially (decreasing) within that time. So, over the ~ 400 ps cooling window, the enhanced diffusion increases the excited spin-packet width by $\approx 3 \mu\text{m}$. The hot-electron phase provides a slight additional contribution to diffusion, but in the grand scheme, the longer, cooler diffusion is the dominant length scale.

In the experiments presented in the main text, the excitation energy was chosen to be close to the probe energy, allowing us to measure the diffusion constant of the cooled electron spins.

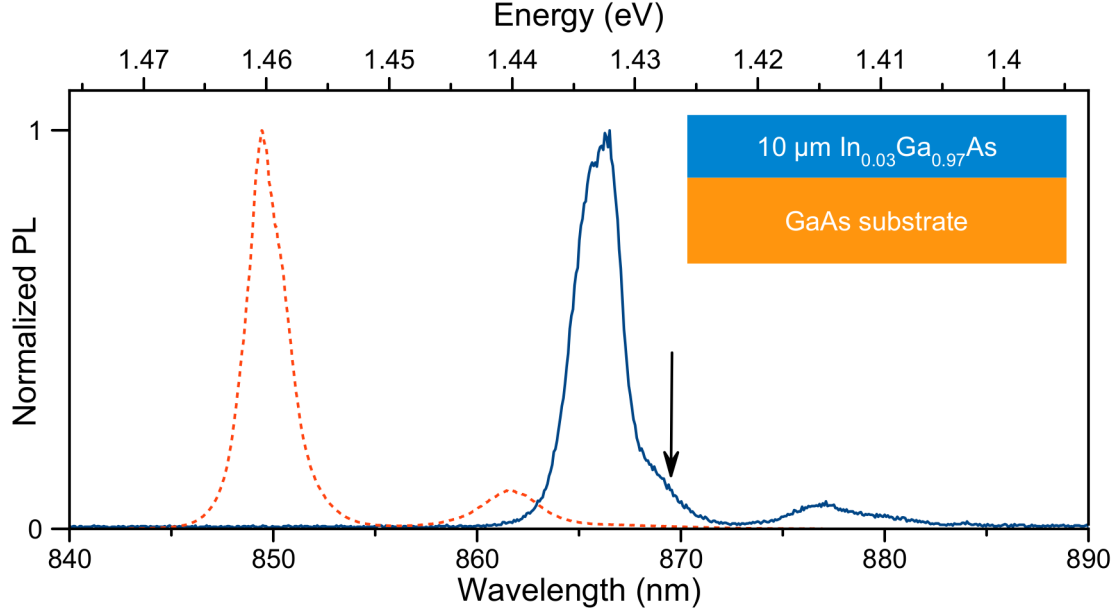


FIG. S1. **PL and Sample.** Normalized PL of two samples. The red dashed line shows the PL of the sample on the thick GaAs substrate. The blue solid line shows the sample with the reduced thickness of the GaAs substrate. The black arrow shows the position of the probe wavelength. Pump is at $E_{\text{exc}} = 1.579 \text{ eV}$ (785 nm), and the probe $E_{\text{pr}} = 1.426 \text{ eV}$ (869.4 nm). The insert shows the sample structure with 10 μm epilayer and GaAs substrate.

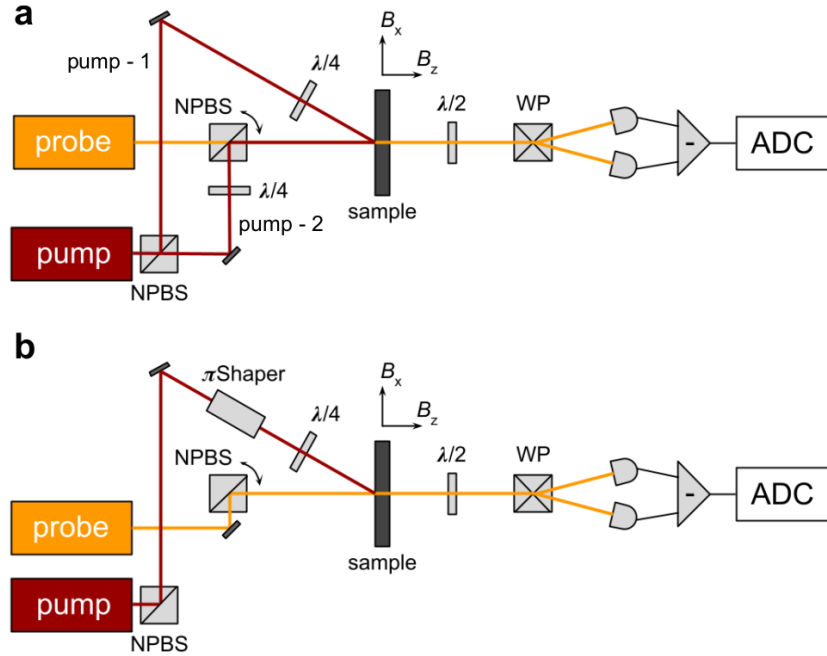


FIG. S2. **Schematics of the experimental setups.** **a**, The setup for the two-pump configuration. The additional pump beam (pump-1, coming under an angle) is blocked for the measurements presented in the main text Fig. 1, and the μm -stages move the cryostat. For the two-pump experiments presented in Fig. 3 of the main text, pump-1 is focused on the center of the probe beam while pump-2 can be shifted at the sample. **b**, Configuration for a flat-top pump measurements, presented in main text Fig. 2. The elements are: a non-polarizing beam splitter (NPBS), $\lambda/2$ and $\lambda/4$ are the half- and quarter-wave plates, a Wollaston prism (WP), and a π Shaper to convert the Gaussian profile to a flat-top beam profile. The rotation of the NPBS in front of the sample was used to shift the spot reflected on the NPBS onto the sample. The energy of the pump laser is $E_{\text{pu}} = 1.579 \text{ eV}$ (785 nm), and of the probe $E_{\text{pr}} = 1.426 \text{ eV}$ (869.4 nm).

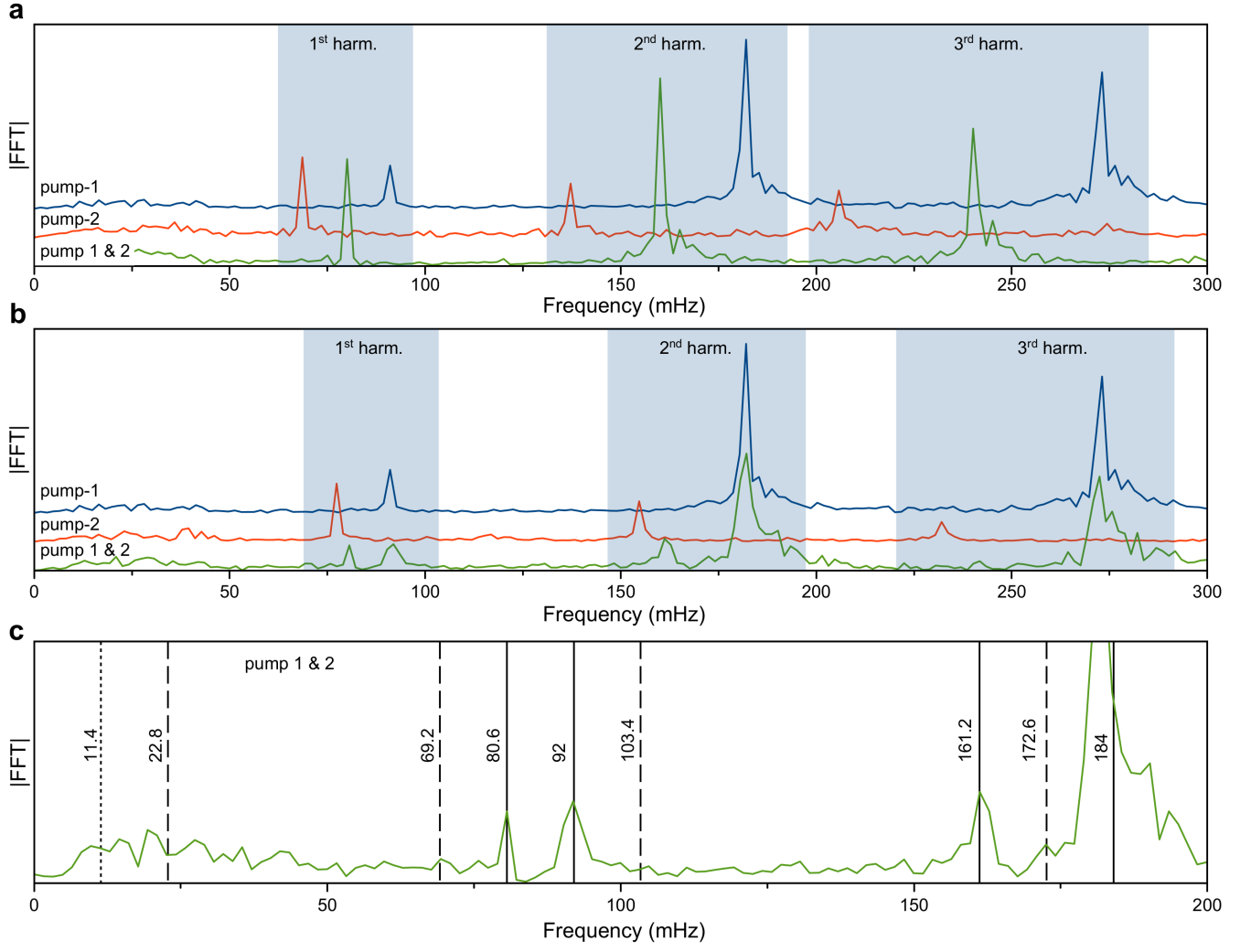


FIG. S3. **Extended FFT data.** **a**, Extended frequency range of the Fig. 3b demonstrating the range of the first three harmonics in the case of synchronization (1:1 synchronization). **b**, Extended frequency range of the Fig. 3e demonstrating the range of the first three harmonics in the absence of synchronization (free-running). **c**, Zoomed-in FFT for the case without synchronization, with the solid lines marking the main frequencies ($f_1, f_2, 2f_1, 2f_2$), a short-dashed line marking the beat (difference), and long-dashed lines marking the intermodulation products with $n, m \leq 2$.

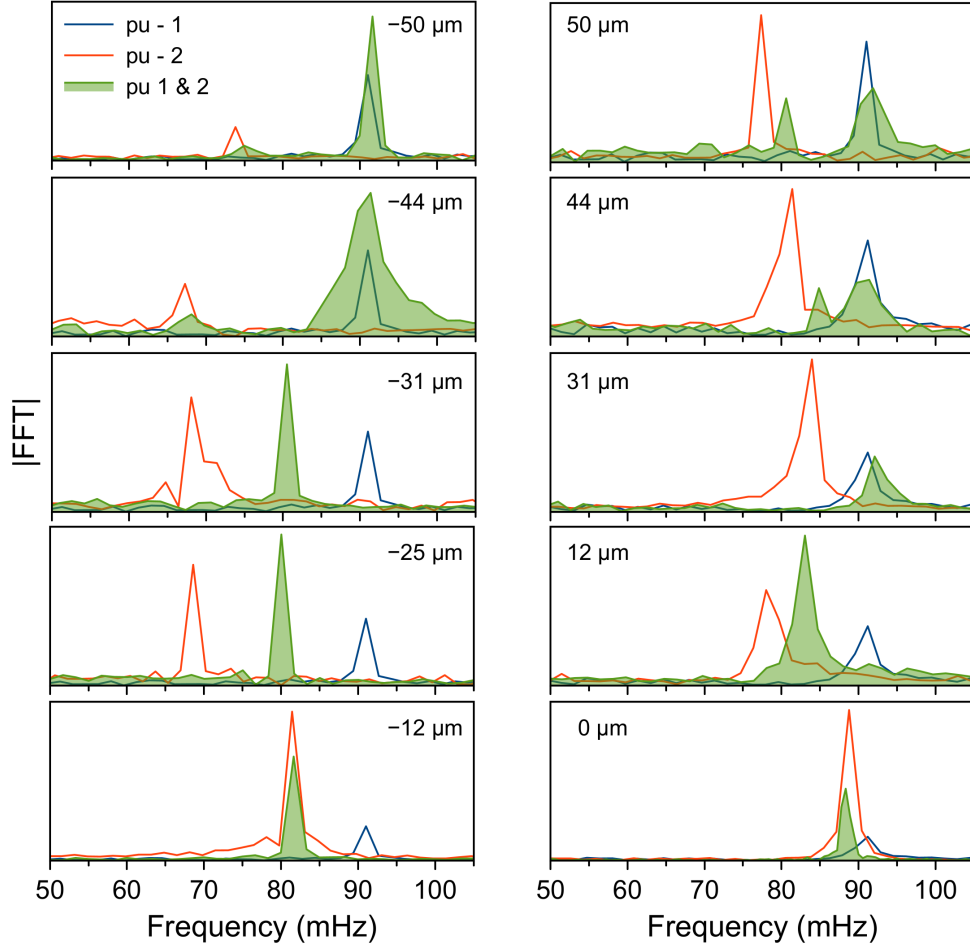


FIG. S4. **FFT at different pump separations.** FFT spectra for pump-1-only (blue), pump-2-only (red), and both pumps together (green shaded) cases for different pump-spot separations.

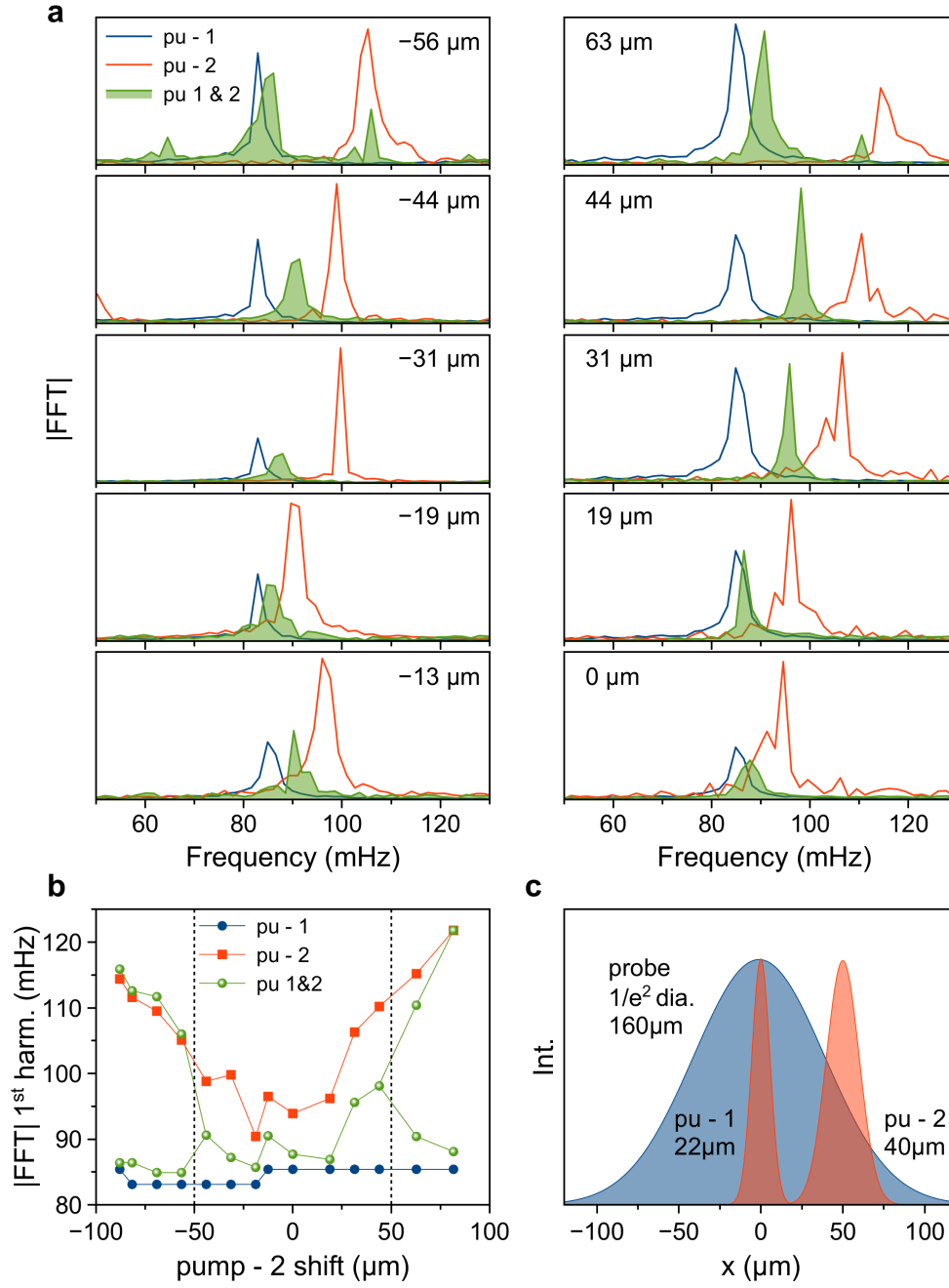


FIG. S5. **Synchronization with spatially wide beams.** **a**, FFT spectra for pump-1-only (blue), pump-2-only (red), and both pumps together (green shaded) cases for different pump-spot separations. **b**, A summarizing plot with points representing the central frequency of the first FFT harmonic. Vertical dashed lines at $\pm 50 \mu\text{m}$ indicate the approximate synchronization border. **c**, Simulation of the measured spot sizes with the probe beam diameter $160 \mu\text{m}$ at $1/e^2$ intensity roll off, blue shaded region. Pump-1 is placed at the center of the probe and has a diameter of $22 \mu\text{m}$, while pump-2 is shifted by $50 \mu\text{m}$ away from pump-1 and has a diameter of $40 \mu\text{m}$.

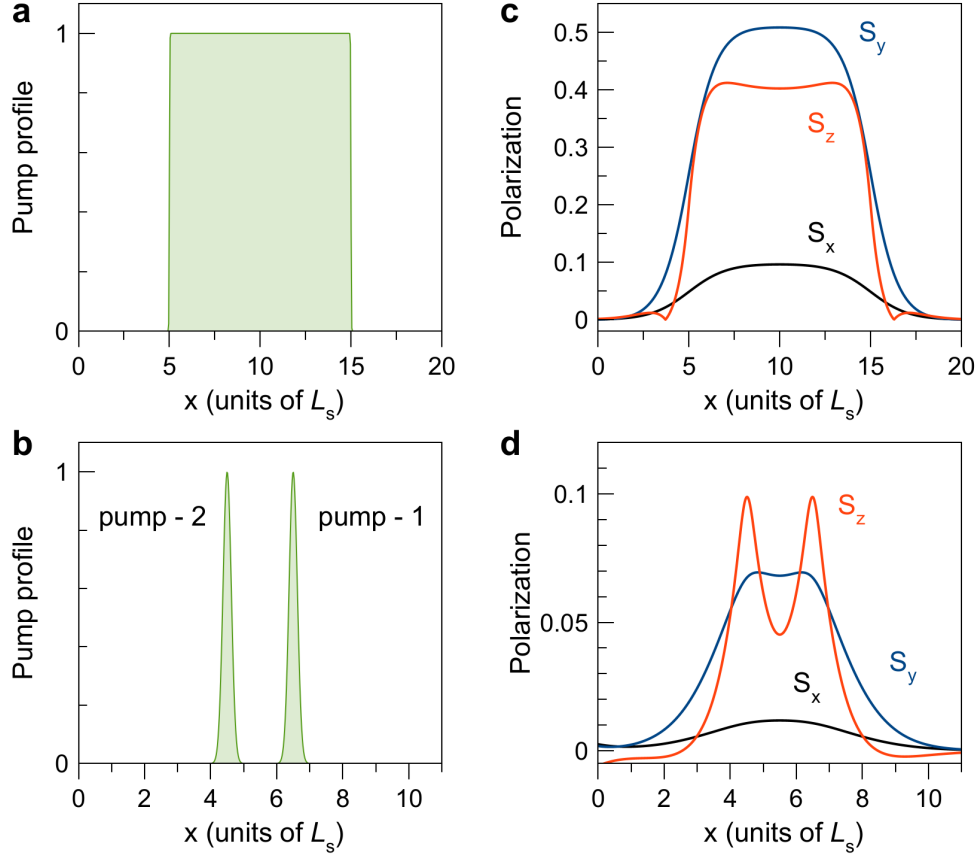


FIG. S6. **Electron spin diffusion in magnetic field.** Spatial profiles of the flat-top pump covering the range from $x = 0$ to $x = 10L_s$ (a) and two Gaussian pumps having FWHM of $0.25L_s$ (b) separated by a distance of $2L_s$. Electron spin polarization components: S_x (black), S_y (blue), and S_z (red), in a magnetic field of $B_x = -1$ mT ($\alpha = 10^\circ$) under excitation by the flat-top pump (c) and by two Gaussian pumps (d).

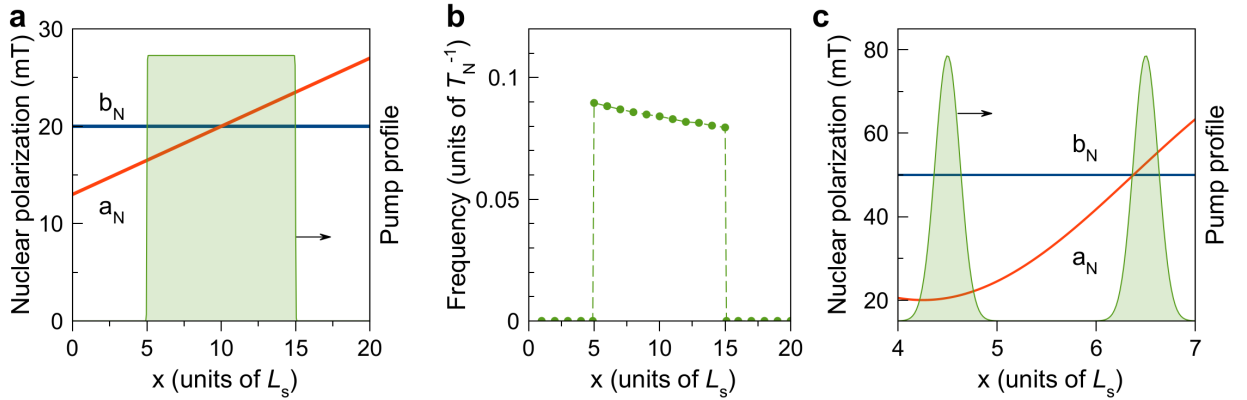


FIG. S7. **Spatial distribution of hyperfine interaction parameters.** a, Spatial dependence of a_N (red) and b_N (blue) under flat-top pump excitation; the green curve indicates the pump profile. b, Spatial dependence of the first harmonic for hyperfine interaction parameters shown in a without spin diffusion. c, Spatial dependence of a_N (red) and b_N (blue) under excitation by two Gaussian pumps separated by $2L_s$; the green curve indicates the pump profiles.

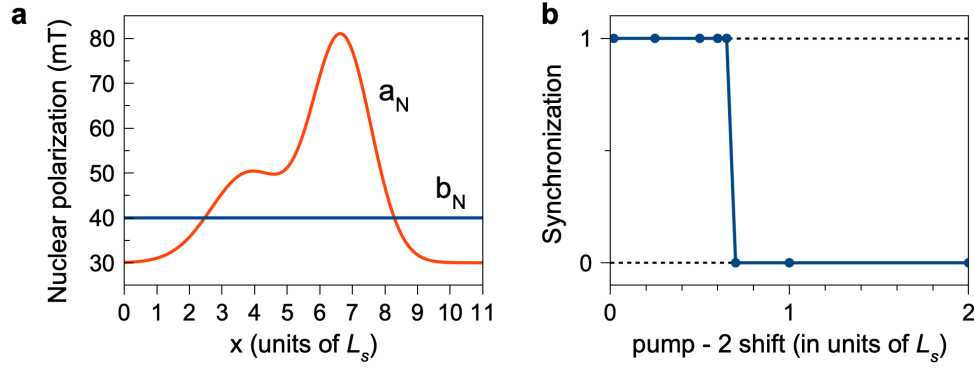


FIG. S8. **Spatial distribution of hyperfine interaction parameters.** **a**, Spatial dependence of a_N (red) and b_N (blue). **b**, Synchronization range for a_N and b_N from panel **a** for pump-2 shifts between 0 and $2L_s$ relative to pump-1.

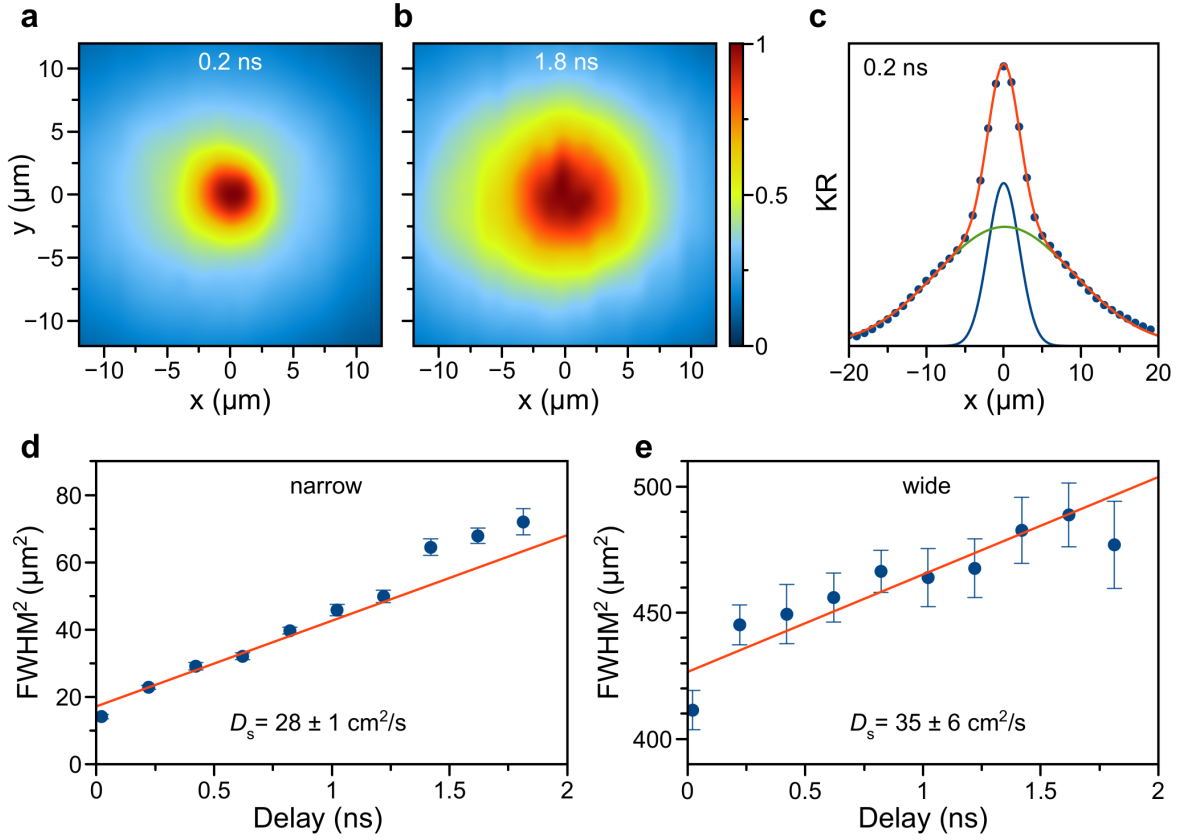


FIG. S9. **Full electron spin diffusion.** Contour plots for the time-resolved pump-probe Kerr rotation experiment with spatial mapping measured at time-delays of 0.2 ns (**a**) and 1.8 ns (**b**), demonstrating the evolution of the spatial profile of the excited spin polarization. The Kerr rotation amplitude is normalized, and its scale is shown by a color bar on the right. **c**, Exemplary double Gaussian fit of the spin polarization at the time delay of 0.2 ns. The blue data points are well-fitted by a narrow (blue line) and a wide (green line) Gaussian profile. The red line is the sum of both fits. **d**, Extracted time evolution of the narrow fit components in FWHM^2 versus time delay. The slope of the linear fit yields the spin diffusion coefficient $D_s = 28 \pm 1 \text{ cm}^2/\text{s}$. **e**, The same for the wide fit component, with $D_s = 35 \pm 6 \text{ cm}^2/\text{s}$.