Strong coupling of alkali-metal spins to noble-gas spins with an hour-long coherence time

R. Shaham 1,2,4, O. Katz 1,2,3,4 and O. Firstenberg 1

Nuclear spins of noble gases can maintain coherence for hours at ambient conditions because they are isolated by complete electron shells. This isolation, however, impedes the ability to manipulate and control them by optical means or by coupling to other spin gases. Here we achieve strong coherent coupling between noble-gas spins and the optically accessible spins of an alkali-metal vapour. The coupling emerges from the coherent accumulation of stochastic spin-exchange collisions. We obtain a coupling strength ten times higher than the decay rate, observe the coherent and periodic exchange of spin excitations between the two gases and demonstrate active control over the coupling by an external magnetic field. This approach could be developed into a fast and efficient interface for noble-gas spins, enabling applications in quantum sensing and information.

Noble-gas isotopes with non-zero nuclear spin, such as helium-3, feature day-long spin lifetimes and hours-long coherence times. They are prominent in various fields, from precision sensing and medical imaging to searches for new physics, and they hold promise for future quantum information applications such as optical quantum memories and the generation of entangled photons. Here we report on strong coherent coupling between the collective spin states of the gas and controlling its quantum excitations.

Polarized ensembles of alkali-metal spins or noble-gas spins can carry such collective excitations, corresponding classically to a tilt of the collective spin about the polarization axis. These can be modelled as quantum excitations of a harmonic oscillator. Remarkably, the quantum description persists even for gaseous ensembles undergoing rapid diffusion and for overlapping ensembles that interact via atomic collisions. The latter rely on the feasibility of preparing the collective spin state of the gas and controlling its quantum excitations.

Polarized ensembles of alkali-metal spins or noble-gas spins can carry such collective excitations, corresponding classically to a tilt of the collective spin about the polarization axis. These can be modelled as quantum excitations of a harmonic oscillator. Remarkably, the quantum description persists even for gaseous ensembles undergoing rapid diffusion and for overlapping ensembles that interact via atomic collisions. The latter rely on the feasibility of preparing the collective spin state of the gas and controlling its quantum excitations.
Here $\omega_a$ and $\omega_b$ denote the Larmor precession frequencies of the collective spins of the alkali-metal and noble-gas atoms, respectively. They are set by the external magnetic field $B$ and by the effective magnetic fields exerted by each species on the other\(^{16}\). We tune $B$ to determine the detuning from resonant coupling $\Delta = \omega_a - \omega_b$. The decoherence rate of the alkali-metal excitations $\gamma$ is included, while for now we neglect the slow decoherence of the noble-gas spins. Finally, $f$ denotes the quantum noise accompanying the relaxation, motion and collision processes\(^{23,24}\). In the current study, $f$ can be discarded, as we prepare the spin ensembles in coherent spin states and study the evolution of the mean transverse amplitudes $\langle \hat{a} \rangle$ and $\langle \hat{b} \rangle$.

\[ \partial_t \begin{pmatrix} \hat{a} \\ \hat{b} \end{pmatrix} = i \begin{pmatrix} \omega_a + i\gamma - J & -J \\ -J & \omega_b \end{pmatrix} \begin{pmatrix} \hat{a} \\ \hat{b} \end{pmatrix} + \hat{f}. \]  

(1)

**Results**

**Experimental setup and protocols.** We study transverse spin excitations of spin-polarized potassium vapour and helium-3 gas enclosed in a spherical glass cell (Fig. 1a). The potassium spins are polarized along the axial magnetic field by an optical pumping beam, and the helium spins are polarized by collisions with the spin-polarized potassium (over 10 h; Extended Data Fig. 1). The cell also contains nitrogen for reducing (quenching) the fluorescence from the optically excited potassium atoms. At low polarization, the helium spins exhibit a coherence time of $T^*_{21} = 2$ h (Fig. 1b), and consequently their individual relaxation is henceforth neglected. The exchange experiments start with turning off the pumping beam.

We monitor the dynamics of the coupled spin system following a short, 5-μs-long pulse of transverse magnetic field $B_z$, which predominantly excites the collective alkali-metal spin and initializes it at a tilt angle of a few degrees from the axial magnetic field $B_z$. We measure the transverse alkali-metal spin using Faraday rotation of an optically detuned, linearly polarized probe beam. In this system, the exchange rate $J$ and the magnetic precession rates $\omega_a$ and $\omega_b$ are all of the same scale when $\Delta \lesssim J$. As a result, strongly coupled dynamics measured in the laboratory frame mixes the effects of Larmor precession with that of the exchange. To eliminate the effect of the former and witness the exchange dynamics directly, we experimentally reconstruct the complex quantities $|\langle \hat{a} \rangle|^2$ and $|\langle \hat{b} \rangle|^2$, each composed of the two spin components (quadratures) in the transverse $x$–$y$ plane. The tomographic-like reconstruction is performed by repeated measurements of the alkali-metal spin dynamics in the $x$–$y$ plane using alternating pulses $B_{1y}$ and $B_{1x}$ for the initial tilt.
calculate the complex amplitude of the collective alkali-metal spin \( \langle a(t) \rangle \). To measure the collective noble-gas spin \( \langle b(t) \rangle \) after some exchange duration \( t \), we halt the exchange dynamics at \( t \) by rapidly ramping up the axial magnetic field (increasing \( \Delta \)) and utilizing the alkali-metal spins as a magnetometer for sensing the noble-gas spin precession.

We realize a maximum coupling rate of \( J = 78 \pm 8 \) Hz by operating at high densities of potassium \( n_k = 4.9 \times 10^{14} \text{cm}^{-3} \) (at \( T = 230 \) °C) and helium \( n_h = 6.45 \times 10^9 \text{cm}^{-3} \) (at room temperature) and with relatively high degrees of spin polarization \( p_c \geq 0.95 \) and \( p_b \geq 0.3 \). At these conditions, collisions among alkali-metal atoms are frequent enough (>0.5 μs\(^{-1}\)) with respect to the Larmor frequency to keep the alkali-metal excitations free from spin-exchange relaxation\(^{22}\). The intricate hyperfine manifold of the alkali-metal atoms maintains a spin-temperature distribution due to these collisions and manifests as an effective spin-1/2 (Fig. 1c)\(^{21}\). Remnant spin relaxation occurring during these collisions dominates the decoherence rate of the alkali-metal excitations \( \gamma = 7.3 \pm 1.5 \) Hz. We thus achieve \( J \geq 10\gamma \). See Methods for a detailed description of the experimental conditions and analysis procedures.

**Dynamics of strongly coupled spins.** Under the strong-coupling conditions, the two spin gases can coherently exchange collective excitations. To demonstrate these dynamics, we tune \( \Delta \) close to resonance and generate an initial excitation predominantly of the alkali-metal spin. Figure 2 presents the measured spin excitations \( \vert \langle a(t) \rangle \vert^2 \) and \( \vert \langle b(t) \rangle \vert^2 \) as they are exchanged back and forth between the two ensembles. Because the magnetic pulse acts also on the noble-gas spin and partially excites it as well, the extinction of \( \vert \langle a(t) \rangle \vert^2 \) at the minima of the observed oscillations is maximized slightly below resonance, at \( \Delta = -1.15\gamma \). The presented measurement is taken at this detuning. This detuning is still small in terms of the strong-coupling dynamics, rendering a near-unity ratio between the exchange and coupling rates \( J/\gamma = 1.15 \), where \( J \approx \sqrt{F^2 + \Delta^2/4} \) is the exchange rate. We observe the reversible exchange and the revival of the excitations back to the alkali-metal spins at \( t = 6.5 \) ms and with a high contrast of 75%, evidencing the strongly coupled dynamics. The uncertainty on the exchange fidelity is small (7.4%) for short exchange times, and it is dominated by uncertainty in the alkali-metal axial polarization (Methods).

As expected, we find that the exchange conserves the total number of excitations \( \vert \langle a(t) \rangle \vert^2 + \vert \langle b(t) \rangle \vert^2 \) aside from the decay introduced by alkali-metal spin decoherence. We also directly observe the slowing down of the exchange oscillations, as the spins gradually decouple due to the dependence of \( J \) on the decaying alkali-metal polarization \( p_c(t) \). The gradual decoupling leads to residual excitations populating the long-lived noble-gas spin. These effects are all captured by a detailed model (solid lines; Methods), which accounts for the temporal decrease of \( J \) and for small geometric misalignments.

**Coupling regimes.** It is instructive at this point to compare the resonant, strong-coupling dynamics with the detuned and overdamped dynamics. These are presented in Fig. 3, showing the measured total number \( \vert \langle a(t) \rangle \vert^2 \) of collective alkali-metal spin excitations (top) and the amplitudes \( \text{Re}(\hat{a}) \) and \( \text{Im}(\hat{a}) \), which exhibit also Larmor precession (bottom). For the coherent spin states in our experiment, oscillations of \( \vert \langle a(t) \rangle \vert^2 \) correspond to nutations (tilt) of the collective alkali-metal spin from the quantization axis \( \hat{z} \) and therefore manifest the exchange interaction in a rotating frame, free of Larmor precession in the \( x-y \) plane.

First, we set \( \Delta \) close to resonance (\( \Delta = -1.15\gamma \) as before) and measure the dynamics under the strong-coupling conditions \( J = 68 \pm 5 \) Hz and \( \gamma = 7.5 \pm 2 \) Hz (Fig. 3a). As in Fig. 2, we observe oscillations of the number of alkali-metal spin excitations \( \vert \langle a(t) \rangle \vert^2 \), exchanged back and forth with the noble-gas spin while gradually decaying. The dynamics far off-resonance is shown for an increased detuning \( \Delta = 460 \) Hz \( \approx 6.8\gamma \) (Fig. 3b). In this regime, we observe a decaying precession of \( \langle \hat{a}(t) \rangle \) and an almost monotonic relaxation of \( \vert \langle a(t) \rangle \vert^2 \) at a rate \( 14 \pm 2 \) Hz, in agreement with the expected value (2\( \gamma \)). Finally, we repeat the experiments with an increased relaxation rate \( \gamma = 215 \) Hz \( \approx 3.2\gamma \) (Fig. 3c), implemented by keeping the pumping beam on during the measurement. The measurements shown
in Fig. 3 of the three regimes elucidate the coherent nature of the exchange interaction under the strong-coupling conditions.

The transition between the overdamped and strong-coupling regimes is continuous, with the reversible dynamics becoming gradually more dominant. At critical damping $f = \gamma/2$, the decay of the alkali-metal spin is shared among both species, and its coherence time is effectively elongated. Figure 3c demonstrates the elongation for on-resonance dynamics (solid) compared with the detuned case (dotted). While $f > 0.5$ promotes an avoided crossing of the normal frequencies of the dynamics (as discussed below), the reversible exchange is negligible at critical damping. At $f = 0.78$, for example, as realized by Kornack and Romalis, only 0.5% of the initial excitations return to the initially tilted gas (Supplementary Section 2). Efficient and reversible exchange of excitations, therefore, requires the ratio $f/\gamma$ to be large.

**Spectral map.** At strong coupling, the system’s response to magnetic fields features a spectral gap. We measure this gap by repeating the experiment presented in Fig. 3a for different values of $\Delta$. The spectral map (Fig. 4a) reveals an avoided crossing between the normal frequencies at $\Delta = 0$, with a wide gap indicating a strong coherent coupling between the two gases. We further compare the measurements with calculated spectra. We present both a simple model based on equation (1) (Fig. 4a, dashed lines) and the results of the detailed model (Fig. 4b,c). Both models reproduce well the main frequency branches. The additional features in the spectrum, primarily the weak perpendicular branches and the vanishing amplitude of the horizontal branch at $\Delta \gtrsim J$ (due to reduced sensitivity to magnetic stimulation near the so-called compensation point), are well captured by the detailed model.

**Discussion**

We realize strong coherent coupling between the collective spins of a dense alkali-metal vapour and a noble gas, with a coupling-to-decay ratio $f/\gamma \approx 10.7$, much larger than unity. The coupling arises from accumulation of stochastic spin-exchange collisions, relying on the weakness of each collision (spin precession of $\sim 10^{-3}$ rad per collision) to conserve coherence and reversibility. We estimate that higher values of $f/\gamma$ are achievable with higher $^3$He density and polarization and at lower temperatures and nitrogen gas pressure. $^3$He pressure exceeding 10 atm was demonstrated as well as 85% polarization. A system at 220°C, with 8.2 atm of $^3$He polarized to 80% and near-unity polarized potassium is expected to reach $f/\gamma > 100$.

Operation of alkali-metal and noble-gas systems in the strong-coupling regime opens several intriguing possibilities. One route motivated by quantum information applications is to use the alkali-metal spins as mediators between photons and noble-gas spins. In particular, fast on-resonance coupling between the spins can enhance the indirect coupling to photons and improve the performance of these applications compared with detuned operation. At strong coupling, read-in, read-out and control of the collective noble-gas spin are done at a rate $f$, whereas detuned operations with $\Delta \gg f$ are done at a substantially lower rate $f/\Delta$ (up to Hz scale). The efficiency and fidelity of the operation are application dependent and could be optimal in either of the two regimes. For long-lived optical quantum memories, the optimal regime depends on the bandwidth $B$ of the optical signal. In systems with $f \gg \gamma$, storage of photons with an optical bandwidth $B \gg k$ (kHz up to GHz) is optimized by first storing the light on the alkali-metal spins and then transferring it to the noble-gas spins via a strong coupling exchange. The exchange efficiency, approximately $\exp(-\gamma t/2)$, approaches unity for $f \gg \gamma$ and could enable hours-long storage with unprecedented time–bandwidth product. Another example relates to the generation of long-lived spin entanglement between multiple cells via detuned operation. Once the entanglement is generated, efficient extraction for subsequent usage requires transfer to the alkali-metal spins, which would rely on strongly coupled exchange. Furthermore, for $f \gg \gamma$, the generation is more efficient regardless of the detuning, as the contribution of alkali-metal projection noise is suppressed by a factor $4\gamma B/f$, where $B^{-1}$ is the duration of the entangling pulse.

A second potential route is to use the strong coupling for improving noble-gas-based sensors. Noble-gas magnetometers sense magnetic fields by measuring the precession of noble-gas spins with an additional auxiliary magnetometer. They are particularly interesting, being fundamentally limited by the low noble-gas projection noise.
Using the alkali-metal spin as an embedded magnetometer enables suppression of its projection-noise contribution by a large factor over the projection noise of the noble gas when $J \gg \sqrt{\gamma/T}$ (ref. 23). In addition, operation in the strong-coupling regime outperforms detuned operations in two aspects. First, detuned operation reduces the magnetic sensitivity at low frequencies near the nuclear magnetic resonance frequency of the noble gas, effectively increasing the impact of photon shot noise. Second, bringing the two spins to a resonance implies lowering the alkali-metal precession frequency, which in turn enables operation in the spin-exchange relaxation-free regime where sensitivity is increased even further24,25. Finally, another application regards self-compensating magnetometers, which typically operate on resonance.26,27. These sensors can readily benefit from an enhanced coupling rate, which could provide for higher bandwidth and dynamic range.

Online content
Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-022-01535-w.

Received: 9 February 2021; Accepted: 4 February 2022;
Published online: 4 April 2022

References

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations. © The Author(s), under exclusive licence to Springer Nature Limited 2022
The polarized spin ensembles exert an equivalent magnetic field on each other, via collisions and via the macroscopic magnetic fields generated by their magnetization. While the equivalent magnetic field experienced by the helium $B_{\text{hel}}=0.98\text{ mG}$ (for $p_s=0.98$) is small, the equivalent magnetic field experienced by the potassium $B_{\text{pot}}=10.94\text{ mG}$ (for $p_s=0.3$) is considerable. The detuning from resonant coupling is thus quite sensitive to $p_s$, which we monitor during the experiment. We do so by applying a constant magnetic field $-B_{\text{hel}}+1.6\text{ mG}$, and monitoring the precession frequency of the decoupled alkali-metal spins following a small transverse magnetic pulse.

In the experiments presented in Figs. 2–4, we use a transverse magnetic pulse to tilt the alkali-metal spins by $\theta = 9.8 \pm 0.2^\circ$, $\theta = 6.8 \pm 0.2^\circ$ and $\theta = 0.7 \pm 0.05^\circ$, respectively. In terms of the number of excitations $|\langle \hat{a} \rangle|^2 \approx q N_p \rho_p^2 J/4$, these correspond to $|\langle \hat{a} \rangle|^2 = (12.1 \pm 0.5) \times 10^{13}$, $|\langle \hat{b} \rangle|^2 = (5.9\pm 0.4) \times 10^{13}$ and $|\langle \hat{a} \rangle|^2 = (6.2 \pm 0.9) \times 10^{11}$.

In all experiments, we measure the transverse spin component of the alkali-metal atoms along the $x$ axis using Faraday rotation of a linearly polarized probe beam. The 5 mm diameter, 260μW probe beam is detuned by $-400\text{ GHz}$ above the D1 transition, and its polarization is measured after the cell using the balanced photodetection method.

We subtract from all measurements a background signal taken without the magnetic pulse. This background signal is small and is dominated by excitations of transverse spins during the fast variation of $B_k$ (when setting $\Delta$) because of the imperfect alignment between the optical and magnetic axes.

**Reconstruction and scaling of $|\langle \hat{a} \rangle\rangle$ and $|\langle \hat{b} \rangle\rangle$.** We use optical Faraday rotation to measure $|\langle \hat{a} \rangle\rangle$ and $|\langle \hat{b} \rangle\rangle$. For the optically broadened line and the far-detuned probe in our setup, and as long as the Faraday rotation angle is small, the balanced-detection readout is proportional to the $x$ component of the collective alkali-metal spin $\langle \hat{S}_x \rangle$, that is, to the electron spin projection along the probing axis. From these measurements, we extract the normalized transverse spin component $\tilde{S}_x(t) = \langle \hat{S}_x \rangle / \langle \hat{S}_z \rangle$. The normalization factor is calibrated separately by tilting the initial spin $\langle \hat{S}_x(0) \rangle$ by all the way to the $x$ direction (equivalent to $\theta = 90^\circ$) and measuring the maximal Faraday rotation angle ($\sim 4$ rad in our system). We verify that the Faraday rotation angle in all subsequent experiments is small.

The measurements of $|\langle \hat{a} \rangle(t)\rangle$ and $|\langle \hat{b} \rangle(t)\rangle$ presented in Fig. 2 are done according to the experimental sequence shown in Extended Data Fig. 3a. The sequence starts by initializing the spins with a small transverse component under conditions of small $\Delta$. After some evolution and partial decay in the dark, at time $t$, we increase $\Delta$ by an order of magnitude (by increasing $B_{\text{hel}}$ to $1.5\text{ mG}$), thus largely decoupling the alkali-metal and noble-gas spins. We continue to monitor the alkali-metal spins and use them as a magnetometer for sensing the noble-gas spins.

During the experiment, while the pumping light is off, the polarization of the alkali-metal spin decays $\rho(t) \neq 0$. This decay changes the slowing-down factor $\theta(t) \neq \theta(p_s)$ and thus shifts the Larmor precession frequency of the alkali-metal spin, which we measure directly (Extended Data Fig. 2a). We model the time dependence of the shift, assuming an exponential polarization decay $\dot{\rho}_x = -\Gamma \rho_x$, where $\Gamma$ is the polarization decay rate at the increased intermediate field. The instantaneous precession frequency of the alkali-metal spin is then given by $\omega_0(\rho_x(t), \Gamma) = 1 + \frac{2}{1 + \frac{p_s}{2}(1+e^{-2\Gamma t})}$, where $\omega_0(\rho_x(t), \Gamma) = 1 + \frac{2}{1 + \frac{p_s}{2}(1+e^{-2\Gamma t})}$.

Here $\sigma(t)$ and $\sigma_0(t)$ are complex fitting parameters, corresponding to the amplitudes of the two frequency components, and $\rho_x, \rho_0, \omega_0, \omega_0, \omega_0, \omega_0$ and $\rho_0$ are real fitting parameters. One such fit is demonstrated in Extended Data Fig. 3b, and the extracted $\rho_x(t), \omega_0(\rho_x, t=0), [\sigma(t)]$ and $[\rho(t)]$ are shown in Extended Data Fig. 2b (note the factor $[\rho(t)]^2 = (\Delta/\bar{\Delta})^2 < 1/1000$. We verify that the extracted amplitudes are insensitive to the exact value of $\Gamma$ (set to $6.8\text{ GHz}$ in all fits) and even to the functional form of $\rho_x(t)$. We use the fits to extract $\rho_x(t)$ (Extended Data Fig. 2b) and find that it is accurately described by the double-exponent function $\rho_x(t) = 0.61e^{-0.0001t} + 0.38e^{-0.01209t}$, presented in Extended Data Fig. 2b with its confidence bounds. The multi-exponential nature of the depolarization can be attributed to multi-mode spatial dynamics of the collective magnetic field $B_k$. The initial magnetic field decay is thus quite sensitive to $\Delta_0$.
\((J\Delta)^2\) and higher. Here we see that the decay of \(p_x(t)\) increases \(\eta\), justifying the relation \(r \propto T\), and explaining the growth of noise in the measurements, which one can interpret as excess thermal excitations. This process leads to the increasing error bars in Fig. 2 at later times. For the experiments presented in Figs. 3 and 4, we reconstruct the complex-valued \(\langle \hat{a}(t) \rangle = \sqrt{n(T)}(\hat{S}_x(t) - i\hat{S}_y(t))\). The two normalized projections \(\hat{S}_x(t)\) and \(\hat{S}_y(t)\) are measured in two consecutive experiments that differ in the direction of the initial pulsed excitation (alternating between \(B_\perp\) and \(B_\parallel\)). In Fig. 2, we average consecutive experiments with the two excitation directions. We use the extracted \(p_x(t)\) for Fig. 3a,b and estimate \(p_y = p_x(0) = 0.98\) for Fig. 3c. Finally, in Fig. 4 we present the normalized Fourier amplitudes \(\langle |\hat{a}(\omega)| \rangle\) as done for the experimental data. In Fig. 4b, we consider a perfectly aligned setup \((\beta_x = \eta_x = \epsilon_x = \epsilon_y = 0)\). In Fig. 4c, we reproduce the imperfection generating the perpendicular frequency branch by introducing a minute misalignment \(\beta_x = 3.1\) mrad and \(\beta_y = -\beta_x\). The calculations for Fig. 2a (solid lines) are done with \(B = 11.33\) mG \((\Delta = -1.15\) and \(B_\perp (t) = 5.2\) mG \(\times 10^{-7}/(2.8\) ms\(^2\)) \((\theta_y = 6.5°)\).

**Data availability**

All data needed to evaluate the conclusions in the paper are present in the paper. The data that support the findings of this study and additional data are available from the corresponding author upon request.

**References**


**Acknowledgements**

We thank C. Arivndak and R. Finkelstein for fruitful discussions. We acknowledge financial support from the Israel Science Foundation, the European Research Council starting investigator grant Q-PHOTONICS 678674, the Minerva Foundation with funding from the Federal German Ministry for Education and Research and the Laboratory in Memory of Leon and Blackye Broder.

**Author contributions**

R.S., O.K. and O.F. all contributed to the experimental design, construction, data collection and analysis of this experiment. R.S. claims responsibility for all figures. The authors wrote the manuscript together.

**Competing interests**

The authors declare no competing interests.

**Additional information**

Extended data is available for this paper at https://doi.org/10.1038/s41567-022-01535-w.

Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41567-022-01535-w.

Correspondence and requests for materials should be addressed to R. Shaham.

Peer review information Nature Physics thanks the anonymous reviewers for their contribution to the peer review of this work.

Reprints and permissions information is available at www.nature.com/reprints.
Extended Data Fig. 1 | Spin-exchange optical pumping. Typical measurement of the pumping process of helium-3 by optically pumped potassium vapor. Here the potassium density is $n_a = 4.9 \cdot 10^{14} \text{ cm}^{-3}$, and the helium depolarization time is $T_{b,\text{act}} = 3.9 \text{ h}$. 
Extended Data Fig. 2 | Variables extracted from fitting Eq. (3) to the measured signals for each exchange time $t$, as exemplified in Extended Data Fig. 3. a. The change in alkali precession frequency $\omega_a(t, \tau = 0)$ [see Eq. (2)] manifests the change in the slowing-down factor due to alkali depolarization. b. The degree of alkali polarization $p_a(t)$ (in semi-log scale). In a and b, dashed black line correspond to the fitted multi-exponential model, and dotted lines present its confidence bounds. These are used as uncertainty estimations when using $p_a(t)$ to scale $\langle \hat{a} \rangle$ and $\langle \hat{b} \rangle$. Less reliable data, extracted when the excitations reside predominantly in the noble gas spins, are marked in gray. c. The two frequency components (amplitude squared) of the normalized Faraday rotation signal $S_x(t + \tau)$. Note the factor of $(\Delta / J)^2 \gtrsim 100$ between them. Each data-point is averaged over 12 to 20 repetitions of the sequence.
Extended Data Fig. 3 | Pulse sequence and typical results of an excitation-exchange measurement. 

**a.** First, we turn off the pumping and bring the two species to strong coupling with a small detuning $\Delta$. We then generate a transverse excitation with a transverse magnetic field pulse. At a later time $t$, we halt the exchange by increasing the axial magnetic field and setting a large $\Delta$. 

**b.** Example of a measured signal with exchange duration $t = 11\, \text{ms}$, with $\Delta = -1.15\, J$ before $t$, and $\Delta = 790\, \text{Hz} \gg J$ after $t$. We measure the alkali electron spin (red) which, once $\Delta$ is increased, can be used as a magnetometer that senses the noble-gas spin. The fast oscillations of the signal correspond to the Larmor precession of the alkali spin, and the slow modulation corresponds to the noble-gas precession. The latter is highlighted by the blue line (generated by low-pass filtering of the signal for illustrative purposes). We fit the signal to the model from Eq. (3) (dashed black line) and find the amplitudes of the alkali and noble-gas components at time $t$, which are used to estimate $\langle a(t) \rangle$ and $\langle b(t) \rangle$, respectively. The same fit also provides $p_a(t)$. 

---

**Extended Data Fig. 3** | Pulse sequence and typical results of an excitation-exchange measurement. 

**a.** First, we turn off the pumping and bring the two species to strong coupling with a small detuning $\Delta$. We then generate a transverse excitation with a transverse magnetic field pulse. At a later time $t$, we halt the exchange by increasing the axial magnetic field and setting a large $\Delta$. 

**b.** Example of a measured signal with exchange duration $t = 11\, \text{ms}$, with $\Delta = -1.15\, J$ before $t$, and $\Delta = 790\, \text{Hz} \gg J$ after $t$. We measure the alkali electron spin (red) which, once $\Delta$ is increased, can be used as a magnetometer that senses the noble-gas spin. The fast oscillations of the signal correspond to the Larmor precession of the alkali spin, and the slow modulation corresponds to the noble-gas precession. The latter is highlighted by the blue line (generated by low-pass filtering of the signal for illustrative purposes). We fit the signal to the model from Eq. (3) (dashed black line) and find the amplitudes of the alkali and noble-gas components at time $t$, which are used to estimate $\langle a(t) \rangle$ and $\langle b(t) \rangle$, respectively. The same fit also provides $p_a(t)$.