

Correlated comagnetometry for precision measurements

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Magnetometers are among the most widely used probes in science and technology. Comagnetometers increase sensitivity by self-cancellation of magnetic noise, but only at low frequencies. We suggest a correlated measurement of two alkali species in one cell to cancel the magnetic background also at high frequencies. The inter-species phase difference of the light-matter interaction response function is found to be calibration free and insensitive to common-mode intensity noise. Utilizing a dark-matter signal as a testcase, the method achieves a thirtyfold background suppression, raising the signal-to-noise ratio by an order of magnitude or more, depending on the coupling to the different subatomic particles. We show that the method also provides model differentiation. The higher sensitivity and model differentiation open a path to novel probes for precision measurements in general and exotic fields in particular.

Optically pumped magnetometers can reach subfemtotesla sensitivity [1, 2] and serve applications from biomagnetism to fundamental physics. In practice, however, the signal-to-noise ratio (SNR) of a shielded magnetometer is rarely set by its intrinsic noise: it is set by the residual magnetic background (shield Johnson noise, coil and current noise), which exceeds the intrinsic floor by more than an order of magnitude [3, 4]. An innermost ferrite shield partially mitigates the Johnson noise passively [3], but leaves coil, current, and other external noise in place. Comagnetometers address this by self-compensation: the polarized noble gas in an alkali-noble-gas cell cancels slow magnetic perturbations [5, 6]. This cancellation, however, is limited to low frequencies, below the noble gas Larmor frequency; above it the device is an ordinary, background-limited magnetometer. In this Letter we introduce a correlated measurement of two alkali species held in a single cell, and theoretically demonstrate that it cancels the correlated (i.e., common-mode) magnetic background at high frequencies as well, recovering the uncorrelated readout-noise floor.

As we will discuss in the outlook, the method lends itself to a variety of applications. In our testcase example below, the correlated background amplitude is suppressed by a factor of ~ 30 , allowing the SNR for a non-magnetic spin interaction to improve by up to an order of magnitude or more, depending on the signal's coupling to the two species. In addition, we simulate a dark-matter transient, and show that the correlated signal also provides a calibration free readout of the signal's coupling structure that no single channel possesses.

Our testcase is built around a dark-matter model involving ultralight pseudoscalar (axionlike) particles (ALPs) [7–9]. Because of the high occupation numbers required to account for the dark matter abundance, they behave as an oscillating classical field. Such fields can

generically couple to the axial-vector currents of Standard Model fermions [10]. In the nonrelativistic limit this acts on nuclear and electronic spins like a magnetic field, but with couplings not proportional to the magnetic moments. Alkali-noble-gas magnetometers and comagnetometers are therefore leading dark-matter detectors [11–15], used to search for oscillations of the field associated with a galactic dark matter halo [13, 16–20], a solar dark matter halo [21], and transients [22, 23]. If such a signal is detected, disentangling its couplings to protons and neutrons has so far required comparing several distinct devices [24]. The correlated single-cell measurement introduced here addresses both needs at once: it removes the magnetic background that limits every single channel, and it reads out the proton-neutron coupling ratio from one vapor cell.

The Dual-Alkali Correlated Measurement. The central observation of this Letter is that two same-cell alkali species in a single comagnetometer provide two independent readouts of the same field environment. ^{87}Rb , ^{39}K , and ^3He are polarized together via hybrid spin-exchange optical pumping [25–27]. The alkalis share the magnetic field and the electronic gyromagnetic ratio γ_e , including the nuclear slowing-down factor. A magnetic field therefore acts on both alkalis via an almost identical coupling. An exotic field, by contrast, couples to each species through its own combination of nucleon couplings, set by the fractional contributions of proton and neutron spins to the total nuclear spin [28]. Because the spin content differs across ^{87}Rb , ^{39}K , and ^3He , one exotic field acts on the three species through three distinct effective couplings, parameterized by the ratio $\mathcal{R} \equiv \xi_n/\xi_p$ of the neutron to proton couplings ($\mathcal{R} = 1$: equal couplings; $\mathcal{R} = 0$: pure proton; any real value is possible in principle).

The common magnetic transfer function makes a differential readout an excellent common-mode rejection filter for the correlated magnetic background, in principle at all frequencies within the linear regime. The cancellation does not rely on the noble-gas self-compensation mechanism [5, 6]; it relies only on the two same-cell alkalis seeing one field. It therefore extends the rejection

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of magnetic noise to frequencies above the compensation point, where the single-alkali comagnetometer no longer attenuates. Consequently the amplitude difference of the two channels, each converted to magnetic-field units, cancels the correlated background itself, converting the background-limited SNR into a readout-noise-limited one, thereby enhancing the sensitivity. A second, complementary observable, is the inter-species phase difference $\Delta\varphi$ which is calibration free and insensitive to common-mode gain and intensity fluctuations, and carries the coupling ratio $\mathcal{R}$. We quantify these observations below.

Experimental Realization. Both observables can be implemented in a standard comagnetometer apparatus (Fig. 1). We take both the magnetic and exotic perturbation fields, $B_\perp$ and $b_\perp$, respectively, to lie along $\hat{y}$. A vapor cell filled with ^3He , ^{39}K , isotopically enriched ^{87}Rb , and N_2 (for quenching) is placed inside a mu-metal shield with magnetic coils, and heated to its operating temperature [29]. A circularly polarized pump laser tuned to the ^{87}Rb D1 resonance polarizes ^{87}Rb directly and ^{39}K and ^3He by spin exchange [27, 30] along $\hat{z}$. The two alkalis are probed simultaneously by two overlapping, linearly polarized beams, each detuned $\sim 0.1\text{ THz}$ to the blue of its own D1 line, traversing the cell along $\hat{x}$. A modulation is added to the probe signal to avoid measuring near the high amplitude region of the $1/f$ noise; the modulator could be a photoelastic modulator [31] or a high-frequency modulation of the shield coils [20], with $\omega_{\text{mod}}/2\pi$ in the tens of kHz, far above the detection bandwidth. After the probes traverse the cell and acquire Faraday rotation, they are split by a narrow-band dichroic beamsplitter followed by an interference filter in each arm, which makes the optical cross-talk between the channels negligible [29], and each polarization is measured by a polarimeter, which is then demodulated by a single multi-channel lock-in amplifier referenced to the shared modulation frequency ω_{mod} , so the two channels share a phase origin and any correlated electronic drift cancels.

For each species $j \in \{^{87}\text{Rb}, ^{39}\text{K}\}$, the in-phase lock-in output returns the Faraday-rotation angle [32],

$$\theta_F^j(t) \propto \ell n_j r_e c f_j \mathcal{D}_j(\nu_j) P_x^j(t), \quad (1)$$

where ℓ is the cell length, n_j the alkali density, r_e the classical electron radius, c the speed of light, f_j the D1 oscillator strength, $\mathcal{D}_j(\nu_j)$ the dispersive lineshape factor at the probe detuning ν_j , and $P_x^j(t)$ the transverse polarization projected onto the probe axis. In a search at an a priori unknown frequency $\omega \ll \omega_{\text{mod}}$, each $\theta_F^j(t)$ is acquired as a time series and Fourier transformed,

$$\tilde{\theta}_F^j(\omega) = \mathcal{F}\{\theta_F^j(t)\} \propto P_x^j(\omega; \mathcal{R}), \quad (2)$$

preserving magnitude and phase. Since the proportionality factor in Eq. (1) is real and positive when the two probes are detuned to the same side of their respective

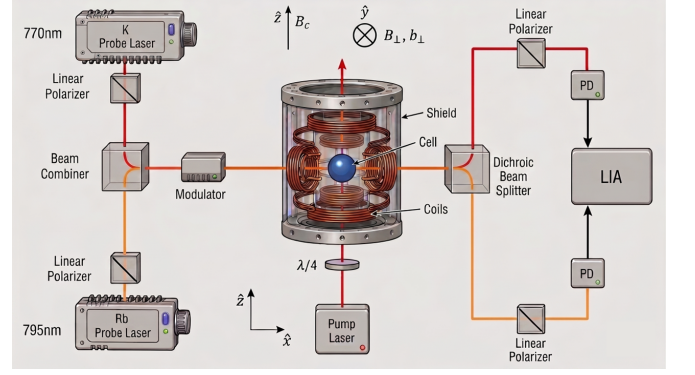


FIG. 1. Experimental setup. A circularly polarized pump polarizes ^{87}Rb along $\hat{z}$ and, via spin exchange, ^{39}K and ^3He . Two overlapping probes near the ^{87}Rb and ^{39}K D1 lines traversing the cell along $\hat{x}$ are separated by a dichroic beam splitter, and each is read out through a polarizer and photodiode (PD), demodulated by a shared lock-in amplifier (LIA) at the modulation frequency. The longitudinal field B_z is tuned by the coils to the compensation point [Eq. (6)]. Both the magnetic and exotic transverse perturbations, $B_\perp$ and $b_\perp$, respectively, point along $\hat{y}$. Auxiliary components are omitted for clarity.

D1 lines (opposite-side detuning adds a known constant π to $\Delta\varphi$), $\varphi_j(\omega; \mathcal{R}) \equiv \arg \tilde{\theta}_F^j = \arg P_x^j(\omega; \mathcal{R})$, and the phase observable is

$$\Delta\varphi(\omega; \mathcal{R}) \equiv \varphi_K(\omega; \mathcal{R}) - \varphi_{\text{Rb}}(\omega; \mathcal{R}). \quad (3)$$

Multiplicative factors in the detected signal that are static over an analysis record (probe intensity, photodetector responsivity, and the atomic prefactor of Eq. (1)) drop out of each phase φ_j , so $\Delta\varphi$ is calibration free.

Coupled Bloch Dynamics. The polarizations $\mathbf{P}_j$ ($j = ^{87}\text{Rb}, ^{39}\text{K}, ^3\text{He}$) evolve under the coupled Bloch equations [14, 24, 33],

$$\begin{aligned} \dot{\mathbf{P}}_j = \frac{1}{q_j} \left\{ \left[\gamma_j (\mathbf{B} + \sum_{k \neq j} \lambda_{jk} M_0^k \mathbf{P}_k) + \xi_j \mathbf{b} \right] \times \mathbf{P}_j \right. \\ \left. + \sum_{k \neq j} \kappa_{jk} n_k (\mathbf{P}_k - \mathbf{P}_j) - R_{pj} (\mathbf{P}_j - \hat{\mathbf{s}}) - \Gamma_j \mathbf{P}_j \right\}, \end{aligned} \quad (4)$$

where γ_j is the gyromagnetic ratio (electronic for the alkalis, nuclear for ^3He), q_j the nuclear slowing-down factor (unity for ^3He), $\mathbf{B}$ the applied magnetic field, $\mathbf{b}$ a transverse exotic pseudomagnetic field with species-specific coupling ξ_j , $\lambda_{jk} M_0^k \mathbf{P}_k$ the Fermi-contact field induced on species j by the magnetization of species k [34], κ_{jk} the spin-exchange rate coefficient, n_k the density of k , R_{pj} the pump rate along $\hat{\mathbf{s}}$, and Γ_j the remaining relaxation. Only ^{87}Rb is pumped; ^{39}K polarizes through the fast ^{87}Rb – ^{39}K spin exchange and both alkalis polarize the ^3He , much more slowly [25]. The exotic couplings are expressed through the nucleon couplings and the nuclear

spin content $\sigma_{n,p}^j$ [28, 35–38] as

$$\xi_j = \eta_j(\sigma_n^j \xi_n + \sigma_p^j \xi_p), \quad (5)$$

where $\eta_j = q_j - 1$ for the alkalis and 1 for the noble gas. Equation (5) assumes a negligible electron-spin coupling; exotic fields coupling to electron spins are also partially screened by magnetic shields, while nucleon couplings are not [39]. Parameter values are provided in Ref. [29].

Equation (4) admits a steady longitudinal solution along $\hat{z}$, set by the pump and the spin-exchange network. In a single-alkali comagnetometer the longitudinal field is tuned to the self-compensation point [5, 6]; with two polarized alkalis this cannot be met for both, since $\lambda_{\text{Rb,He}} \neq \lambda_{\text{K,He}}$, so we parameterize the compensation field as a weighted compromise,

$$B_z(\alpha) = -[(1 - \alpha)\lambda_{\text{Rb,He}} + \alpha\lambda_{\text{K,He}}]M_0^{\text{He}}P_z^{\text{He}} - \lambda_{\text{Rb,He}}M_0^{\text{Rb}}P_z^{\text{Rb}} - \lambda_{\text{K,He}}M_0^{\text{K}}P_z^{\text{K}}, \quad (6)$$

with $\alpha = 0$ ($\alpha = 1$) recovering ^{87}Rb -optimal (^{39}K -optimal) compensation. The fast ^{87}Rb – ^{39}K spin exchange locks the alkalis into a single collective transverse mode, fixing the optimal α near the ^{39}K fraction of the alkali density. We adopt $\alpha = 0.578$ for our cell [29]. Together with the cell parameters it also sets the antiresonance location below. The transverse linear response at frequency ω is evaluated numerically following the fast scheme of Ref. [24].

Figure 2 presents the physics of the correlated dual-alkali measurement. Panels (a) and (b) contrast the ^{39}K response of the dual-alkali cell with a reference ^{39}K – ^3He single-alkali cell. Both attenuate magnetic drives at low frequency, but they differ sharply for an exotic drive: for $\mathcal{R} = 0$ the dual cell develops an antiresonance in the response $|P_x^K|$ near 157 Hz that the ^{39}K – ^3He cell does not have at this coupling ratio. Quite generally, with $\xi_j = \eta_j(\sigma_n^j \xi_n + \sigma_p^j \xi_p)$ and $\eta_j > 0$, an indirect drive opposes the direct one only when the two species’ couplings differ in sign, i.e., when $\mathcal{R}$ lies between their sign-reversal ratios $-\sigma_p^j/\sigma_n^j$. At $\mathcal{R} = 0$ this excludes ^{39}K – ^3He (^{39}K and ^3He share coupling sign) but not ^{87}Rb – ^{39}K – ^3He , where ^{87}Rb supplies the opposing arm. The values of $\mathcal{R}$ for which each alkali–noble pair supports an antiresonance are tabulated in Ref. [29]. Panel (c) resolves the ^{39}K response into its three exotic channels: the direct drive, the ^3He back-action (whose Fermi-contact return fails to cancel because $\lambda_{\text{Rb,He}} \neq \lambda_{\text{K,He}}$), and the alkali–alkali spin exchange. Their in-phase cancellation produces the antiresonance dip. The dip becomes an exact null on a one-dimensional locus in $(\alpha, \mathcal{R}, \omega)$ where the two real conditions $|P_+| = |P_-|$ and $\arg[P_+/(P_-)^*] = \pi$ hold simultaneously [29]; the $\alpha = 0.773$ curve in panel (b) sits on this locus at 72 Hz. Panel (d) shows the resulting phase anatomy: each alkali’s phase winds through its own antiresonance in opposite senses, so the difference $\Delta\varphi$ accumulates a single sharp swing, while for a

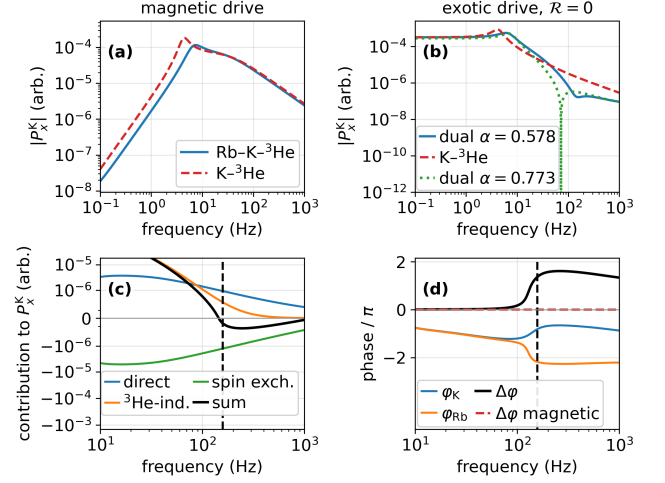


FIG. 2. Physics of the correlated dual-alkali response. (a),(b) Absolute steady-state transverse response $|P_x^K|$ to a perturbation along $\hat{y}$. (a) Magnetic drive: the dual-alkali ^{87}Rb – ^{39}K – ^3He cell (solid blue) and a reference ^{39}K – ^3He single-alkali cell (red dashed) both show the standard low-frequency self-compensation. (b) Exotic drive with $\mathcal{R} = 0$: self-compensation no longer holds at low frequency, and the dual-alkali cell develops a sharp antiresonance near 157 Hz at $\alpha = 0.578$ (solid blue). Changing only α to 0.773 (green dotted; a B_z change of 2.6 nT, i.e., 1.3%) moves it to ≈ 72 Hz, onto the perfect-cancellation locus where $|P_x^K|$ nulls completely [29]. The ^{39}K – ^3He single-alkali reference (red dashed) has no antiresonance at this coupling ratio [29]. (c) Mechanism at $\alpha = 0.578$, $\mathcal{R} = 0$: the three contributions to the ^{39}K response: the direct exotic drive (blue), the ^3He -induced field (orange), and the alkali–alkali spin exchange (green), shown as complex phasors projected onto the direct-drive phasor. Near the dip (dashed line) the opposing spin-exchange term cancels the direct-plus- ^3He drive and the in-phase sum (black) crosses zero; the quadrature component keeps the dip a finite minimum. (d) Phases φ_K (blue), φ_{Rb} (orange), and $\Delta\varphi$ (black) for the exotic drive: each alkali’s phase winds rapidly through its own antiresonance, in opposite senses, so $\Delta\varphi$ accumulates a single sharp swing. Crucially, neither feature appears in the magnetic drive: panel (a) shows no antiresonance in the response, and panel (d) shows a small $\Delta\varphi$ (red dashed) at the milliradian level across the band [29].

magnetic drive $\Delta\varphi$ stays at the milliradian level across the band. The location of the structure depends on $\mathcal{R}$ but also strongly on α , providing an experimental handle through B_z .

Dual-Channel Noise Cancellation. We now use the dual channels to cancel the correlated magnetic background and quantify the SNR gain. The starting point is the Fourier-transformed lock-in signal of each channel, $\hat{\theta}_F^j(\omega)$ [Eq. (2)]. We convert each channel to magnetic-field units by dividing out its magnetic transfer function $T_j^B(\omega)$, the complex response of $\hat{\theta}_F^j$ to a known applied field, measured once by driving a calibrated field with the system’s coils and forming $T_j^B = \hat{\theta}_F^j/\tilde{B}$ at each fre-

quency:

$$x_j(\omega) \equiv \frac{\tilde{\theta}_F^j(\omega)}{T_j^B(\omega)} = B(\omega) + \chi_j(\omega) b(\omega) + \frac{N_j(\omega)}{T_j^B(\omega)}, \quad (7)$$

with $\chi_j \equiv T_j^b/T_j^B$ the exotic response in field units and N_j the uncorrelated readout noise. The common optical prefactor of Eq. (1) multiplies $\tilde{\theta}_F^j$ and T_j^B alike and cancels in the ratio, so no absolute optical calibration is needed and the true field B enters both channels with coefficient one. The correlated background is removed by an adaptive subtraction $r = x_K - \hat{H} x_{Rb}$, where $\hat{H}(\omega) = \langle x_K x_{Rb}^* \rangle / \langle |x_{Rb}|^2 \rangle$ is the least-squares (Wiener) transfer from the ^{87}Rb to the ^{39}K channel, a spectral ratio read off exotic-signal-free records rather than a fitted model [29]. Because $\hat{H}$ is measured, the cancellation is self-calibrating: any error in the transfer functions T_j^B folds into $\hat{H}$ and cancels, entering only the amplitude scale of the recovered signal. The residual $r = (\chi_K - \chi_{Rb})b$ plus uncorrelated noise retains the exotic signal with $g(\omega) \equiv |\chi_K - \chi_{Rb}|/|\chi_K|$ relative to the single ^{39}K channel, while the correlated background cancels to the uncorrelated readout floor; the waveform is then recovered by Wiener deconvolution in a declared search band [29].

Figure 3 illustrates the scheme on a simulated dark-matter transient, an ALP-star crossing [15, 40] in which the axion field oscillates at its Compton frequency while the transit sets the envelope, modeled here as a carrier $f_0 = 100$ Hz under a Gaussian envelope of 0.25 s full width at half maximum, injected into a realistic background: shield Johnson noise at $7 \text{ fT}/\sqrt{\text{Hz}}$ with a $1/f$ rise below 20 Hz [3, 4], an uncorrelated readout floor of $0.17 \text{ fT}/\sqrt{\text{Hz}}$ per channel [1], and light-shift and spin-projection noise within the budget of Ref. [29]. Throughout, the noise floor denotes the rms amplitude spectral density over the declared 70–120 Hz search band (excluding $f_0 \pm 5$ Hz), and the SNR is the signal’s spectral amplitude at f_0 divided by this floor. In the single ^{39}K channel the transient is buried, at SNR 0.5 [Fig. 3(a)]; the dual-channel subtraction cancels the correlated background, and the transient emerges in the denoised record at SNR 7.3 [Fig. 3(b)], an improvement of $\times 15$. The waveform itself is then recovered by Wiener deconvolution of the residual [29]. The amplitude spectral density shows the mechanism [Fig. 3(c)]: the noise floor in the search band drops from 6.2 to $0.22 \text{ fT}/\sqrt{\text{Hz}}$, a factor of 28, down to the dual-channel readout floor, while the 100 Hz signal survives. Figure 3(d) generalizes the simulation across the band. Because the background suppression $S(f)$ and the signal retention $g(f)$ are frequency dependent, the net SNR gain grows with frequency across the band, reaching $\times 15$ at 100 Hz and peaking at $\approx \times 30$ between the ^{87}Rb and ^{39}K exotic antiresonances, where the ^{87}Rb exotic response χ_{Rb} dips and the retention exceeds unity. At low frequencies, the referred exotic responses χ_j con-

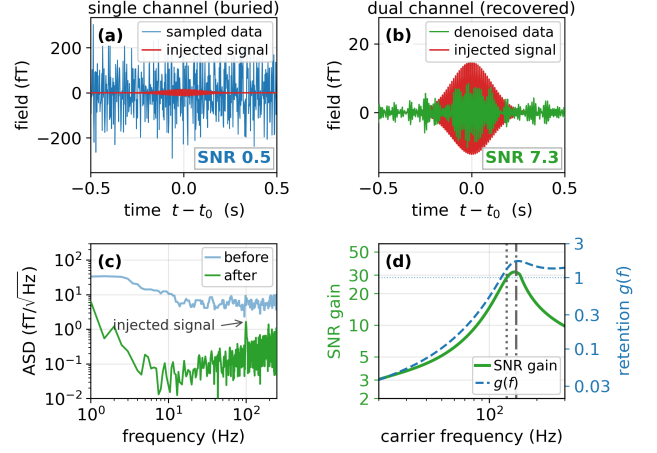


FIG. 3. Two-channel noise cancellation on a simulated dark-matter transient (carrier $f_0 = 100$ Hz, Gaussian envelope, FWHM 0.25 s, $\mathcal{R} = 0$); 2 s sampled at 500 Hz, in magnetic-field units. (a) Single-channel (^{39}K) simulation (blue) with the injected transient overlaid at true scale (red, 15 fT peak): buried at SNR 0.5. (b) ^{39}K channel after dual-beam noise cancellation, band-pass filtered to the search band as in an actual search, with the injected transient again overlaid (red); the transient emerges at SNR 7.3, a $\times 15$ improvement, its recovered amplitude reduced by the retention g . The SNRs are spectral quantities, fixed by the noise-floor drop and the signal retention $g \simeq 0.53$, independent of this time-domain display band-pass. (c) Amplitude spectral densities of the full records in (a) and (b): the floor near f_0 drops from 6.2 (correlated noise) to $0.22 \text{ fT}/\sqrt{\text{Hz}}$ ($\times 28$, the two-channel readout floor), while the 100 Hz signal peak survives (arrow). (d) Amplitude SNR gain over the more sensitive single channel for a transient as in (a), as a function of its carrier frequency f_0 (green, left axis), and signal retention $g(f)$ (blue dashed, right axis); the gain peaks between the ^{87}Rb and ^{39}K antiresonances of the field-referred exotic response χ_j [Eq. (7)] (dotted and dash-dotted verticals, 129 and 148 Hz).

verge ($g \rightarrow 0$); the scheme targets the band above the detection bandwidth, where the single cell is otherwise background-limited. The suppression $S(f)$ is coupling independent and scales with the assumed correlated-to-uncorrelated noise ratio; the retention g depends on $\mathcal{R}$, with the simulated $\mathcal{R} = 0$ lying within the coupling-ratio ranges predicted by the canonical axion model classes discussed below [29].

Coupling-Ratio Readout. Beyond detection, the dual channel measures a property of the signal that no single channel carries: the neutron-to-proton coupling ratio $\mathcal{R}$. Across the band the exotic phase difference $\Delta\varphi(\omega; \mathcal{R})$ exceeds the milliradian magnetic baseline by up to three orders of magnitude and varies steeply with $\mathcal{R}$: at 100 Hz it moves by hundreds of milliradians between the benchmark ratios of the canonical model classes discussed below (-0.17 rad at $\mathcal{R} = 0.04$, $+0.13$ rad at $\mathcal{R} = -0.46$), so comparing the measured cross-spectral phase differ-

ence to a reference map determines $\mathcal{R}$ [29]. The map itself need not rely on the model calculation: a synthetic exotic field, formed by combining species-selective light shifts with a magnetic field [24], generates $\Delta\varphi(\omega; \mathcal{R})$ in situ, making the readout end-to-end calibration free. The measured ratio then confronts theory. The two canonical axion model classes predict overlapping ranges: KSVZ-type models fix $\mathcal{R} = 0.04(6)$, while DFSZ-type models sweep $\mathcal{R}$ monotonically across $[-0.41, +0.65]$ as $\tan\beta$ traverses its perturbative window, broadening to $\approx[-0.47, +0.88]$ once the coupling uncertainties are included [7, 41, 42]. A measured $\mathcal{R}$ therefore pins down the coupling structure that any candidate model must reproduce, rather than naming the model itself: within the DFSZ class it determines $\tan\beta$; a value incompatible with 0.04(6) excludes the KSVZ class; a value outside $\approx[-0.47, +0.88]$ excludes both, pointing to a generic ALP. The axion–electron coupling present in these models can manifest in two ways. Acting directly on the electron spins, it drives the two alkalis identically and leaves ^3He untouched, so the amplitude subtraction rejects it as common mode; a field induced by the shield’s electron-spin response [39] carries the magnetic signature and is likewise rejected. In the phase channel the residual effect is negligible for KSVZ, vanishes for DFSZ-II at large $\tan\beta$ (though not at small), and for DFSZ-I modifies the phase signature at the tens-of-percent level across the band, scaled by an $\mathcal{O}(1)$ shield-response factor, so a DFSZ-I interpretation should fit the full coupling set.

A single-channel spin-based sensor is sensitive to only one effective coupling and cannot separate the proton and neutron contributions; recovering their ratio has so far required comparing several distinct devices [24]. In contrast, the calibration-free, dual-channel method can enable a measurement of the underlying coupling structure even with a single vapor cell.

Discussion. The correlated dual-alkali measurement adds four capabilities to a conventional comagnetometer: it subtracts the correlated magnetic background to the uncorrelated readout floor—a ~ 30 -fold suppression that raises the SNR by up to more than an order of magnitude (depending on coupling)—in the high-frequency band where self-compensation fails; it recognizes an exotic perturbation by an inter-species phase difference up to three orders of magnitude above the magnetic baseline; it supports a tunable antiresonance that can be brought to exact nulling; and it reads out the coupling ratio $\mathcal{R}$ calibration free from a single cell, so that for a sufficiently

strong detection the same data determine the signal’s nucleon coupling structure, the quantity against which candidate models are tested, information a single spin-based sensor does not carry. The antiresonance, like the phase map, can be generated and verified in situ with the synthetic exotic field [24]. The scheme applies to any signal that couples to the two species differently than a magnetic field does, and requires only modest additions to a standard search apparatus (a second probe laser, its polarimetry chain, and a dichroic splitter), allowing retrofits of existing devices.

Outlook. The scheme is not specific to exotic fields: any perturbation that acts on the two species differently than a magnetic field does is converted into a differential signal, while the magnetic background is subtracted. This opens several directions. In principle, an electron electric dipole moment, whose atomic enhancement differs strongly between ^{87}Rb and ^{39}K , becomes a species-selective signal, while the magnetic noise of the electrode currents is subtracted as common mode. Dual light-shift spectroscopy at selected wavelengths can determine ratios of dipole matrix elements with high precision. The compensation optimum and the antiresonance location depend on the alkali– ^3He Fermi-contact enhancement factors, offering a single-cell precision measurement of their ratio, currently known only to a few percent [26]. Extending the method to more alkalis with comb-referenced probes would overdetermine the couplings while suppressing differential probe drifts. This single-cell correlated approach opens a path to high-sensitivity probes of exotic fields and a new generation of detectors for precision measurements.

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SUPPLEMENTAL MATERIAL

1. Simulation parameters and nuclear spin content

This section collects the numerical inputs used throughout the paper. Table I lists the cell parameters, rates, and field settings that enter the coupled Bloch equations [Eq. (4) in the main text] for the dual-alkali ^{87}Rb - ^{39}K - ^3He system. Table II provides the fractional contributions of the neutron and proton spins to the total nuclear spin of each species; these spin-content factors $\sigma_{n,p}^j$ determine the species-dependent exotic couplings ξ_j via Eq. (5) in the main text. Table III lists, for each alkali-noble pair, the range of the coupling ratio $\mathcal{R} \equiv \xi_n/\xi_p$ over which an exotic-field antiresonance can occur. The cell uses isotopically enriched ^{87}Rb ; natural rubidium is 72% ^{85}Rb ($I = 5/2$), and enrichment leaves a single rubidium isotope, so that the ^{87}Rb channel has a single, well-defined slowing-down factor.

TABLE I. Simulation parameters for the dual-alkali ^{87}Rb - ^{39}K - ^3He comagnetometer. The ^3He gyromagnetic ratio and magnetic moment are negative; magnitudes are listed. The alkali vapor densities follow from the saturated-vapor-pressure formulas [32] scaled by the 10:90 ^{87}Rb : ^{39}K mole fraction.

Parameter	Value	Units
Temperature, T	180	$^{\circ}\text{C}$
^3He concentration, $n_{^3\text{He}}$	3	amg
^{87}Rb - ^3He spin-exchange rate coefficient, $\kappa_{\text{Rb,He}}$	6.7×10^{-20} [26]	cm^3/s
^{39}K - ^3He spin-exchange rate coefficient, $\kappa_{\text{K,He}}$	5.5×10^{-20} [26]	cm^3/s
^{87}Rb - ^{39}K spin-exchange rate coefficient, $\kappa_{\text{Rb,K}}$	1.2×10^{-9} [26]	cm^3/s
^{87}Rb - ^3He enhancement factor, $\lambda_{\text{Rb,He}}$	$8\pi/3 \times 6.2$ [26]	
^{39}K - ^3He enhancement factor, $\lambda_{\text{K,He}}$	$8\pi/3 \times 5.8$ [26]	
Electron gyromagnetic ratio, γ_e	$2\pi \cdot 2.8$	MHz/G
^3He gyromagnetic ratio, $\gamma_{^3\text{He}}$	$2\pi \cdot 3.24$	kHz/G
Alkali magnetic moment, μ_{alk}	9.274×10^{-21}	erg/G
^3He magnetic moment, $\mu_{^3\text{He}}$	$1.159 \times 10^{-3} \mu_{\text{alk}}$	erg/G
^{87}Rb relaxation rate, Γ_{Rb}	600	Hz
^{39}K relaxation rate, Γ_{K}	600	Hz
^3He relaxation rate, $\Gamma_{^3\text{He}}$	5×10^{-5}	Hz
^{87}Rb pump rate (dual-alkali cell), $R_{p,\text{Rb}}$	1200	Hz
^{39}K pump rate (^{39}K - ^3He cell), $R_{p,\text{K}}$	600	Hz
Compensation parameter, α	0.578	
^{87}Rb : ^{39}K ratio (mole fraction)	10:90	%
^{87}Rb vapor density, n_{Rb}	4.0×10^{13}	cm^{-3}
^{39}K vapor density, n_{K}	5.45×10^{13}	cm^{-3}
^{87}Rb equilibrium polarization, P_z^{Rb}	0.46	
^{39}K equilibrium polarization, P_z^{K}	0.45	
^3He equilibrium polarization, P_z^{He}	0.047	

2. The compensation optimum α^*

The compensation field of Eq. (6) in the main text interpolates only the ^3He -on-alkali term between the Rb-optimal ($\alpha = 0$) and K-optimal ($\alpha = 1$) settings, because a single applied B_z cannot simultaneously compensate both alkalis when $\lambda_{\text{Rb,He}} \neq \lambda_{\text{K,He}}$. The optimal weight has a simple physical origin. The ^{87}Rb - ^{39}K spin-exchange rate is large (Table I), so the two alkalis are locked into a single collective transverse mode and precess together. This collective spin experiences the ^3He magnetization not as two separate fields but as one effective field equal to the number-weighted average of the individual couplings,

$$\langle \lambda_{A,\text{He}} \rangle = \frac{n_{\text{Rb}} \lambda_{\text{Rb,He}} + n_{\text{K}} \lambda_{\text{K,He}}}{n_{\text{Rb}} + n_{\text{K}}}, \quad (8)$$

TABLE II. Fractional contributions of the neutron and proton spins, σ_n^j and σ_p^j , to the total nuclear spin of each species. ^3He , ^{39}K and ^{87}Rb are the species of the dual-alkali cell analyzed in this work; ^{21}Ne is included as a further noble gas for the general comparison of Table III. The ^3He values rest on a full-scale shell-model calculation, the ^{39}K values on a detailed perturbation-theory calculation, and the ^{87}Rb and ^{21}Ne values on the corresponding nuclear-structure estimates; see the cited references. These spin-content factors set the species-dependent exotic couplings ξ_j through Eq. (5) in the main text.

Species	σ_n	σ_p	Refs.
^3He	0.87	-0.027	[28, 35]
^{21}Ne	0.196	0.013	[36]
^{39}K	0.034	-0.131	[28, 37]
^{87}Rb	0.083	0.251	[38]

TABLE III. Range of $\mathcal{R}$ for which the read-out alkali A and the noble gas N have opposite-sign exotic couplings. With $C_j = \sigma_p^j + \mathcal{R}\sigma_n^j$, the condition $C_A C_N < 0$ is necessary for an exotic-field antiresonance; it holds for $\mathcal{R}$ between the sign-reversal ratios $\mathcal{R}_j^* = -\sigma_p^j/\sigma_n^j$ when σ_n^A and σ_n^N share a sign (as for all pairs here), and outside that interval when they differ. In the dual-alkali ^{87}Rb - ^{39}K - ^3He cell ^{39}K has both partners, so its range is the union $(-3.02, 3.85)$, which contains $\mathcal{R} = 0$.

Pair (A - N)	$(\mathcal{R}_A^*, \mathcal{R}_N^*)$	Antiresonance range
^{39}K - ^3He	(3.85, 0.03)	$0.03 < \mathcal{R} < 3.85$
^{87}Rb - ^3He	(-3.02, 0.03)	$-3.02 < \mathcal{R} < 0.03$
^{39}K - ^{21}Ne	(3.85, -0.07)	$-0.07 < \mathcal{R} < 3.85$
^{87}Rb - ^{21}Ne	(-3.02, -0.07)	$-3.02 < \mathcal{R} < -0.07$

where the weights are the polarized-spin populations of the two alkalis (equal here, since $P_z^{\text{Rb}} \simeq P_z^{\text{K}}$, so the number densities suffice). Matching the interpolated ^3He term $[(1 - \alpha)\lambda_{\text{Rb,He}} + \alpha\lambda_{\text{K,He}}]$ to this collective coupling gives

$$\alpha^* = \frac{n_{\text{K}}}{n_{\text{Rb}} + n_{\text{K}}} \approx 0.576, \quad (9)$$

the ^{39}K fraction of the total alkali density. For the cell of Table I, Eq. (9) evaluates to 0.576, and the value that numerically minimizes the residual very-low-frequency (DC) magnetic response is 0.579 for the ^{39}K channel (0.570 for ^{87}Rb), bracketing Eq. (9). At this weight a DC magnetic field is suppressed by $\sim 1.4 \times 10^4$ in the ^{39}K channel and $\sim 2.7 \times 10^3$ in the ^{87}Rb channel relative to the on-resonance response—one to two orders of magnitude better than the ~ 150 obtained when either alkali is set to its own compensation point ($\alpha = 0$ or 1), because the compromise matches the locked collective mode rather than either alkali individually. We adopt $\alpha = 0.578$ throughout.

Compensation nulls the DC *amplitude* response, but the two alkalis keep a small difference in Larmor frequency, which leaves a residual magnetic phase difference $\Delta\varphi^B$ that grows toward low frequency: about 2.5 mrad at 100 Hz, 15 mrad at 10 Hz, and larger toward DC. The phase-based rejection therefore stays at the milliradian level within the 70–120 Hz search band considered here, and degrades below it; equalizing the two Larmor frequencies with a species-selective light shift would extend it to lower frequencies.

3. The exact-null locus of the antiresonance

The transverse polarization is decomposed in the Bloch ansatz $P_\perp(t) = P_+ e^{i\omega t} + P_- e^{-i\omega t}$, with P_+ and P_- obtained from the linearized steady state. The x -projection is $P_x = [P_+ + (P_-)^*]/2$, which is proportional to the observable [Eq. (2) of the main text], and vanishes exactly when $P_+ = -(P_-)^*$. This is a single complex condition, equivalent to two real ones: amplitude equality, $|P_+| = |(P_-)^*|$, and a phase offset of π between P_+ and $(P_-)^*$. These constitute two real equations on the three-parameter space $(\alpha, \mathcal{R}, \omega)$, so the subset on which both hold simultaneously is one-dimensional, and along it $|P_x^K|$ is exactly zero. The operating points shown in Fig. 2(b) of the main text illustrate the sharp difference between antiresonances that do ($\alpha = 0.773$, 72 Hz) and do not ($\alpha = 0.578$, 157 Hz) belong to this subset. The same mechanism produces an analogous antiresonance in the ^{87}Rb response at a different frequency (≈ 130 Hz at $\alpha = 0.578$, $\mathcal{R} = 0$), which is exploited by the amplitude channel in Fig. 3(d) of the main text.

4. The phase-difference map and recovery of the coupling ratio

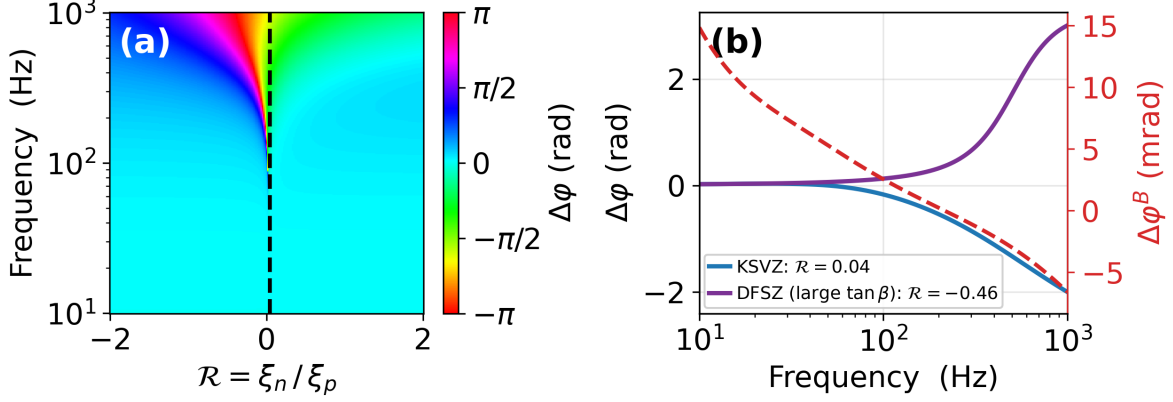


FIG. 4. $\Delta\varphi(\omega; \mathcal{R})$ for the dual-alkali Rb-K- ^3He cell at $\alpha = 0.578$. (a) Heatmap of $\Delta\varphi$ across the $(\mathcal{R}, \omega)$ plane, wrapped to $[-\pi, \pi]$ for the cyclic color bar; the black dashed line marks the central value of the KSVZ prediction, $\mathcal{R} = 0.04$. (b) Unwrapped cross sections at two representative model-motivated ratios, $\mathcal{R} = 0.04$ (central KSVZ prediction, blue) and $\mathcal{R} = -0.46$ (large- $\tan\beta$ DFSZ boundary, purple), comparing the exotic phase difference $\Delta\varphi$ (left axis, radians) to the magnetic baseline $\Delta\varphi^B$ (dashed red, right axis, milliradians): a separation of up to three orders of magnitude enabling high common-mode rejection. Beyond the antiresonance $|\Delta\varphi|$ exceeds π , so the unwrapped curves in (b) equal the wrapped map in (a) up to 2π . The map suppresses the magnetic response to the milliradian level while serving as a matched filter for a specific coupling ratio $\mathcal{R}$.

Figure 4 shows how to recover $\mathcal{R}$ by mapping $\Delta\varphi$ across the $(\omega, \mathcal{R})$ plane [panel (a)] and contrasts the exotic and magnetic responses along representative cross sections [panel (b)]: for the model benchmarks the exotic phase difference exceeds the magnetic baseline $\Delta\varphi^B$ by up to three orders of magnitude across the band, while $\Delta\varphi^B$ itself remains negligible across the band. Given a candidate transient that triggers the system, one computes the cross-spectral phase between the simultaneously recorded $P_x^K(t)$ and $P_x^{\text{Rb}}(t)$ records over the frequency band and compares it to the calibration map $\Delta\varphi(\omega; \mathcal{R})$ for $\mathcal{R}$.

The nuclear spin-content factors $\sigma_{n,p}^j$ carry theory uncertainties: at the percent level in absolute terms for ^3He and larger for the alkalis. For the recovered $\mathcal{R}$ this translates to a comparable fractional uncertainty. The sign-reversal ratio $\mathcal{R}_{\text{He}}^* = -\sigma_p^{\text{He}}/\sigma_n^{\text{He}} = 0.027/0.87 \approx +0.03$ is more sensitive, since σ_p^{He} is itself a small number: a percent-level absolute shift moves $\mathcal{R}_{\text{He}}^*$ by tens of percent, shifting the nucleon ratio to which the measured antiresonance and benchmark phases correspond. These spin-content uncertainties enter the recovered $\mathcal{R}$ on the same footing as the theory uncertainties of the model predictions themselves: KSVZ-type models fix $\mathcal{R} = 0.04 \pm 0.06$, dominated by the nucleon coupling $C_n = -0.02(3)$, with $C_p = -0.47(3)$ [41], while DFSZ-type models sweep $\mathcal{R}$ monotonically downward across $[-0.41, +0.65]$ as $\tan\beta$ (defined as v_u/v_d) traverses its perturbative window $0.25 \leq \tan\beta \leq 170$, broadening to $\approx [-0.47, +0.88]$ once the coupling uncertainties are included, and passing through the KSVZ band near $\tan\beta \approx 0.74$. The observable is therefore reported as a measured coupling ratio: it excludes a model class only when the value falls outside that class's range, and it cannot separate the classes where their predictions coincide.

Cross-talk between the two probe channels contributes a small systematic to $\Delta\varphi$. Optical leakage of one probe into the other's photodiode is suppressed to the 10^{-6} level by a narrow-band interference filter in each polarimetry arm. What remains is atomic: each probe picks up a small Faraday rotation from the other alkali's far dispersive tail, at the percent level or below for probes detuned ~ 0.1 THz from their own D1 lines, which translates to a $\Delta\varphi$ offset of a few mrad. This offset is static and is absorbed by the in-situ synthetic-field calibration. A strong light-shift transient might appear as an exotic-field candidate: it does not share the magnetic channel ratio, so it survives the correlated subtraction and shows up in the residual r just as a genuine signal would. The second observable, the phase difference, separates the two. A light shift couples to the alkalis species-selectively rather than through the common electronic gyromagnetic ratio, so it arrives with its own phase signature, set by the ratio of the shifts it imposes on the two alkalis. This ratio is fixed by the laser detunings and does not depend on the amplitude of the transient. In the Bloch model at 100 Hz, a shift acting on Rb (K) alone produces $\Delta\varphi = -66$ (+51) mrad, compared with +2.5 mrad for the magnetic background and -0.17 (+0.13) rad for an exotic drive at $\mathcal{R} = 0.04$ (-0.46). A candidate whose phase difference falls off the calibrated map $\Delta\varphi(\omega; \mathcal{R})$ is therefore identified as a light-shift artifact and rejected, at no cost

to genuine events.

5. Two-channel cancellation and Wiener recovery

The field-referred channels of Eq. (7) in the main text, $x_j = \tilde{\theta}_F^j/T_j^B$, are combined as $r(\omega) = x_K(\omega) - \hat{H}(\omega) x_{\text{Rb}}(\omega)$, with the transfer estimate

$$\hat{H}(\omega) = \frac{\langle x_K(\omega) x_{\text{Rb}}^*(\omega) \rangle}{\langle |x_{\text{Rb}}(\omega)|^2 \rangle} \quad (10)$$

computed by Welch averaging over exotic-signal-free records (the magnetic background is always present and is precisely what $\hat{H}$ learns); for transient searches these are the off-trigger records, while for a persistent (continuous) signal $\hat{H}$ must be estimated from neighboring frequency bins to avoid partial self-nulling of the signal. The quality of $\hat{H}$ is set by the number of averaged segments rather than by any single window: in the band where the correlated background dominates the referred channels, x_j , the ordinary coherence between them is near unity, so the cross-spectral estimator converges within a handful of segments (seconds of data at the simulation sampling), and the useful record length is limited by the stationarity of the background rather than by counting statistics. The simulation estimates $\hat{H}$ from a 200 s signal-free record (100 two-second segments). Since the correlated background dominates both referred channels, $\hat{H} \rightarrow 1$, and any miscalibration of the magnetic transfer functions T_j^B folds into $\hat{H}$ and cancels; the model enters only the amplitude scale of the recovered waveform. The exotic waveform is then recovered by Wiener deconvolution in the declared search band,

$$\hat{b}(\omega) = \frac{G^*(\omega) r(\omega)}{|G(\omega)|^2 + S_{nn}(\omega)/S_b(\omega)}, \quad G \equiv \chi_K - \hat{H} \chi_{\text{Rb}}, \quad (11)$$

where S_{nn} is the residual (uncorrelated) noise spectrum and S_b a flat prior for the signal spectrum in the band; no template for the signal's shape or timing is assumed. The simulation of Fig. 3 in the main text uses a sampling rate of 500 Hz, a declared search band of 70–120 Hz, and a 2 s spectral window, with the transient amplitude set so that the single-channel spectral SNR is 0.5.

The search itself is broadband: the subtraction and the retention $g(f)$ operate across the declared band at a fixed operating point, with no scanning, and the usual per-bin trials factor of a spectral search applies. The phase readout requires no tuning: the cross-spectral phase of the recorded data is compared to the map at the known operating point α . It is possible to choose the operating point in advance to place the steep region of the map within the search band; for persistent candidates, stepping B_z additionally tests whether the signal tracks the calculated α dependence.

6. Noise budget for the dual-channel subtraction

The adaptive subtraction removes any perturbation that carries a common magnetic signature; the residual floor is therefore set by noise that is not common-mode. Four classes are relevant.

Species-selective (light-shift) noise. Pump light-shift fluctuations, driven by drifts in probe intensity, detuning, or beam pointing, act on a single alkali as an effective transverse field, the same signature class as the exotic signal. The spin-exchange locking that pins the magnetic phase difference also transmits a ^{87}Rb -only effective field to the ^{39}K channel with amplitude ratio 0.975 at 100 Hz; accounting for the small relative phase of the two field-referred transfers, the subtraction suppresses it by ~ 14 , leaving $\approx 7\%$. The exotic signal is not suppressed in the same way because part of it is carried by a ^3He -mediated arm that a light shift lacks. An effective-field drive appears in the field-referred channel with transfer $|T_{\text{Rb}}^{\text{LS}}/T_{\text{Rb}}^B| \approx 0.43$ at 100 Hz; requiring the residual to fall below the recovered $0.22 \text{ fT}/\sqrt{\text{Hz}}$ floor then sets a budget line $\delta B_{\text{LS}} \lesssim 7 \text{ fT}/\sqrt{\text{Hz}}$ of ^{87}Rb -equivalent effective field at 100 Hz, met by standard detuning and intensity control. The simulation of Fig. 3 injects $1 \text{ fT}/\sqrt{\text{Hz}}$, well within this budget.

Response-ratio stability. The cancellation holds while the ratio $c \equiv T_K^B/T_{\text{Rb}}^B$ of the two magnetic transfer functions is stable. A fractional drift δc in this ratio leaks the correlated background into the residual r at the level δc , so reaching the $0.22 \text{ fT}/\sqrt{\text{Hz}}$ floor requires $\delta c \lesssim 3\%$ over the timescale on which $\hat{H}$ is re-estimated: This is a stability requirement, not a calibration: $\hat{H}$ measures the ratio from the data, so its absolute value is never required; it must only stay constant between successive re-estimations. This requirement is loose: because spin-exchange locking makes the two alkalis respond as a single collective mode, c is nearly insensitive to the operating point—in the Bloch model, $d \ln |c|/dT \approx 5 \times 10^{-4} \text{ K}^{-1}$ at 100 Hz, so ordinary cell-temperature control leaves orders of magnitude of margin, and $\hat{H}$

is in any case re-estimated from each ~ 200 s record. The operating point itself, and with it the antiresonance location and the phase map, follows the ^3He magnetization through the compensation condition. Closed-loop control of the compensation field, demonstrated on the same ^{39}K – ^{87}Rb – ^3He platform in Ref. [30], holds this point fixed against the slow (hours-scale) ^3He polarization dynamics, so the phase map needs only occasional re-verification with the synthetic-field calibration rather than continuous re-measurement.

Differential gradient sampling. ^{87}Rb is pumped directly while ^{39}K is polarized through spin exchange, so the two polarization profiles are not identical, the two channels sample the field at slightly different effective positions, and a magnetic-field gradient is not perfectly common-mode. The offset between the two effective positions is bounded by the distance a K atom diffuses before equilibrating with the local Rb polarization, $\ell = \sqrt{qD/(k_{\text{RbK}} n_{\text{Rb}})} \approx 50 \mu\text{m}$ for the parameters of Table I, so the two profiles coincide to this scale. Even allowing a tenfold margin (0.5 mm), shield Johnson-noise gradients at the level of Ref. [3] contribute at most $0.08 \text{ fT}/\sqrt{\text{Hz}}$, a fraction of the residual floor.

Spin-projection noise. The standard single-species estimate, $\delta B \approx (1/\gamma_e)\sqrt{2q\Gamma/(n_j V)}$ [1, 5], with the densities of Table I and the effective volume $V \approx 1 \text{ cm}^3$ over which the 10 mm probe beam overlaps the pumped column in our 20-mm-diameter spherical cell, gives 0.06 and $0.07 \text{ fT}/\sqrt{\text{Hz}}$ for the ^{39}K and ^{87}Rb channels; added in quadrature by the subtraction, this is about $0.09 \text{ fT}/\sqrt{\text{Hz}}$, a factor of two below the readout floor.

All four error budget terms lie at or below the recovered $0.22 \text{ fT}/\sqrt{\text{Hz}}$ readout floor, so none limits the simulation of Fig. 3 in the main text.

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