



OPEN Parametrising harmonic strengths in Raman driven spin noise spectra of neutral atoms

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We study the appearance of harmonics in spin noise correlations as a result of strong optical drive using a pair of coherent Raman beams in hot Rubidium vapour. In addition to the fundamental spin noise mode, we detect multiple equi-spaced coherence signals, including first, second, third, and fourth harmonics with relative amplitudes ranging from 10^{-1} to 10^{-4} . We parametrise these higher harmonics with Raman and probe beam intensity, density of atoms, magnetic field strength and drive frequency. These observations allow us to precisely estimate the energy spacing between magnetic sub-levels in a multi-level atomic system under the influence of external magnetic fields. Our findings highlight this method as a versatile tool for optical probing and precision metrology of complex spin dynamics.

Keywords Polarised light, Spin noise spectroscopy, Quantum sensing, Multi-photon optical drive

Driven spin dynamics in atomic vapours provide a rich setting to explore coherence, relaxation, and correlations in multi-level quantum systems. Among the available tools, spin noise spectroscopy (SNS)^{1–6} is a method for studying the spin dynamics of a system by measuring the magnetisation fluctuations of an ensemble of atoms at thermal equilibrium. Here, one measures fluctuations in Faraday rotation angles of a weak, far-detuned probe beam. This non-invasive detection scheme makes it a useful candidate for atomic magnetometry used in quantum sensing^{7,8}, biomedical imaging⁹, archaeological surveys¹⁰, space exploration¹¹, and navigation¹². While equilibrium SNS have been broadly used and already reveal features such as spin-alignment peaks at twice the Larmor frequency¹³, externally driving the system in and out of equilibrium can amplify these signals, and it reveals valuable information about the driven system, which was otherwise obscured in the linear response equilibrium spin noise signals. It generates higher harmonics and higher-order correlators as observed in driven semiconductor SNS¹⁴ and two-photon SNS in alkali atoms¹⁵. More interestingly, when the drive is strong, there is higher harmonic formation in this driven atomic system. This method, involving the interplay between light and spin fluctuations, reveals new insights into probing spin dynamics and magnetic properties in materials¹⁶. To reveal complex magnetic phenomena and expand our understanding of non-equilibrium, multi-level spin systems, Raman driven harmonic generation holds the capability of uncovering possible long-range coherences in the ground-state Zeeman manifold. Another advantage of this system is that it could be extended to study spin correlations in more complex multi-electron atoms like Helium, as well as semiconductors and solid-state systems where additional resonance peaks may arise due to higher-order electron–electron (spin–spin) correlations^{17–19}. In this study, we experimentally investigate Raman driven SNS in hot vapours of ⁸⁵Rb atoms. By applying two coherent optical fields, we generate effective couplings between ground-state Zeeman sublevels of the atom. Using dispersive Faraday rotation measurements, we observe higher harmonics of the Larmor frequency (ν_L) in the spin noise spectrum. We characterize the strengths of the harmonics empirically with respect to various controllable parameters, such as probe beam intensity, coherent drive strength and frequency, density of atoms, and magnetic field strength, which will serve as a basis for developing a detailed theoretical description²⁰. In multi-level atomic systems, the application of bichromatic Raman beams leads to multiple electromagnetically induced transparency (EIT) resonances that connect Zeeman or hyperfine sublevels, with amplitudes and positions determined by selection rules and magnetic field geometry²¹. Additionally, pulse-modulated magnetic drive of spin ensembles has been shown to restructure the spin-noise spectrum, shifting it to fractional multiples of the modulation frequency and uncovering otherwise hidden dynamics²². Moreover, coherent population trapping in multi- Λ configurations produces rich spectra where additional lasers connect dark states, demonstrating how multi-path interference between coupled levels can enhance nonlinear responses²³. Despite these advances, a systematic framework connecting SNS to the rich physics of harmonic

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generation is still missing. Our driving scheme of Raman driven SNS (RDSNS) provides a platform for exploring harmonic-like behaviour of the fundamental spin-noise signal, but with a narrower linewidth in spin ensembles as a result of the interplay of driven oscillation and the inherent spin fluctuations. This approach provides a powerful tool for probing non-equilibrium multi-level spin systems and opens new avenues for precision metrology in complex atomic and correlated systems.

Observation of multiple equispaced RDSNS peaks

The basic experimental setup, which we previously reported in²⁴, was used for precision magnetometry. It comprises two main laser sources (Probe and Raman) derived from two separate External Cavity Diode Lasers (ECDLs) centred at 780 nm (Toptica Photonics DL100). The probe laser is blue detuned by 31 GHz from $^2S_{1/2}$ $F = 3$ to $^2P_{3/2}$ $F = 4$ transition line of ^{85}Rb . It is made linearly polarised before entering the Vapour cell set up with a polarisation purity greater than 99.99 %. The probe light is further focused at the centre of the cell using a lens combination such that the focal waist is 45 μm . After coming out of the vapour cell, the probe beam is collimated again and sent to a polarising beam splitter (PBS122), which splits the light equally from the two ports. The light from two ports of the PBS falls on a balanced photodetector (see Fig. 1a) having a bandwidth of 100 MHz (Thorlabs, PDB230A). To get the power spectrum of the signal, we further connect the photodetector output to a Spectrum analyser (Keysight, N1996A). A second ECDL, designated as the Raman laser, is kept on resonance with $^2S_{1/2}$ $F = 3$ to $^2P_{3/2}$ $F = 4$ transition line of ^{85}Rb . The Raman laser output is divided into two beams, which are passed through Acousto-Optic Modulators (AOMs) (ISOMET 1206C and AA OPTICS MT110-A1-IR) in separate double-pass configurations for precise frequency and intensity control. The beam sizes were made identical (waist diameter = 2.2 mm) by suitable lens combinations. Care was taken to make the Raman beams have circular (σ^+) and linear (Π) polarisation with a polarisation purity of above 99.99%. They were merged using a non-polarising beam splitter (BS014). Light from one of the output ports of the NPBS was sent through the vapour cell set-up. Light from the other output port was used to monitor the frequency difference (δ_{12}) between the two Raman beams by detecting the beat frequency of the interfering Raman beams using a photodetector (Thorlabs, DET36A) and an oscilloscope. The merged Raman beams were made to counter-propagate with respect to the probe beam propagation direction (see Fig. 1a). This was to ensure that no part of the stray Raman beam falls on the detector. The absolute frequencies of all the light sources were monitored using a wavelength meter (High Finesse GmbH WSU) with a frequency error of ± 1 MHz, along with saturation

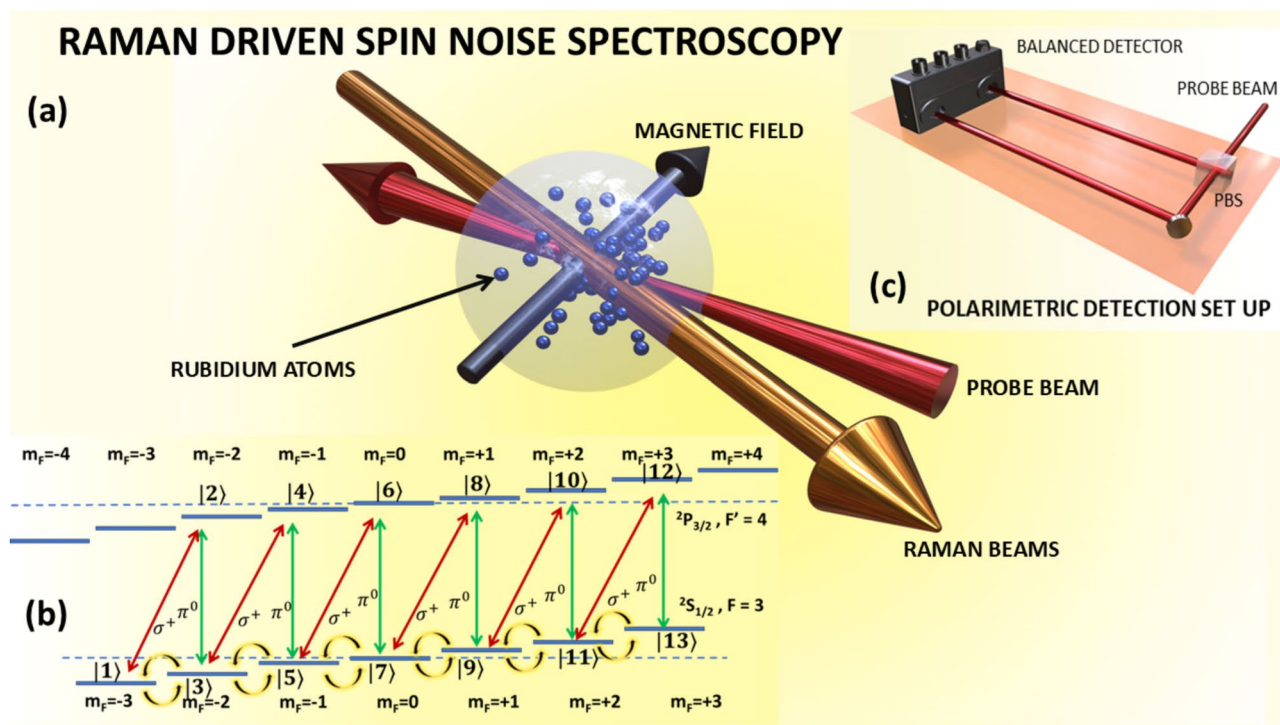


Fig. 1. Experimental set up. (a) A linearly polarised probe beam (red) is focused at the centre of a Rubidium vapour cell. A pair of coherent beams (yellow) (Raman beams) with particular polarisations (σ^+ and π) is merged and made counter-propagating with respect to the probe beam (red). The magnetic field (black) is kept perpendicular to the probe and Raman beams propagation direction. (b) Scheme of the Experiment. The pair of Raman beams connects the magnetic states of the ground hyperfine Zeeman manifold. Due to the multi-level structure, the atoms are driven to succeeding levels. The excited states are represented by even ket notations, while the ground states are represented by odd ket notations. (c) Illustrates the polarimetric detection setup. The polarisation fluctuation of the probe beam is measured using a PBS (polarising beam splitter) and a balanced photo-detector.

absorption spectroscopy setups. The vapour cell used for this experiment is cylindrical in shape and made up of Pyrex, having a length of 50 mm and a diameter of 25 mm. It contains both the isotopes of Rubidium in natural abundance. In order to reduce the effect of spin-destructive collisions, which broaden the linewidth, the cell is filled with a buffer gas of Neon at 200 torr. To control the density of atoms, we heat the cell using strip heaters (OMEGA KHLVA-105/10-P) and a PID temperature controller (OMRON E5CWL). The set-up is thermally insulated using ceramic blankets. We maintain a temperature stability of $\pm 0.05^\circ$. A uniform magnetic field is created in the direction perpendicular to both the probe and Raman beams propagation direction using a pair of coils in a Helmholtz configuration. We used a stable current controller (LD3000, Thorlabs) for the magnetic coil with a stability of $< 1 \mu A$. The maximum spatial variation of the field was measured to be 2 parts in 10^5 over the detection region of 4.5 mm. The temporal variation of the generated field was limited to less than ± 0.1 nT over 1 sec of measurement time. To shield the setup from external stray magnetic fields, it was enclosed in a four-layer μ metal casing, which shields the field by a factor of 10^4 times. In the Raman driven power spectrum, the atoms are driven between the magnetic states (m_F) of the ground hyperfine level via magnetic sub-levels of the excited state by using a pair of laser beams as shown in Fig. 1. The Hamiltonian of the system can be described as

$$\begin{aligned} \frac{H}{\hbar} = & \sum_i \omega_i^e |i\rangle \langle i| + \sum_j \omega_j^g |j\rangle \langle j| + \Omega_1 \sum_{\{m,n\}} (|m\rangle \langle n| e^{-i\omega_{s1}t} + |n\rangle \langle m| e^{i\omega_{s1}t}) \\ & + \Omega_2 \sum_{\{m,n\}} (|m\rangle \langle n| e^{-i\omega_{s2}t} + |n\rangle \langle m| e^{i\omega_{s2}t}) \end{aligned} \quad (1)$$

where ω_i is the angular frequency of state $|i\rangle$ and the superscript e or g represents the excited and the ground state. Here, Ω_1 and Ω_2 are the resonant Rabi angular frequencies of Raman field 1 (R_1) and Raman field 2 (R_2), respectively. The states following the selection rule allowed for electric dipole transition is denoted by $\{m, n\}$.

For a simplified 3 level atomic system, for example, the one constituting the ground states $|1\rangle$ and $|2\rangle$ and the excited state $|3\rangle$, there is negligible decay between the states $|1\rangle$ and $|2\rangle$ compared to decay rates from $|3\rangle$ to $|1\rangle$ and $|2\rangle$, we have chosen the following limits on the relaxation rates: $\gamma_{13} = \gamma_{23} = \gamma$, $\gamma_{12} = \gamma'_{21} = \gamma'$, and $\gamma' \ll \gamma$. Here, γ' is the spin coherence relaxation rate between the two Zeeman states, and the relevant correlation function is the auto-correlation of the ground-state coherence $\rho_{21} = \langle 1|\rho|2\rangle$. The steady-state expression for ρ_{21} in the Raman resonance limit ($\Delta = 0$, $\delta_{12} = \nu_L$) has been derived in Ref. ²⁵. For the 3 level case, we note that the power spectrum looks like a Dirac delta peak with an envelope as $|\rho_{21}|^2$ which grows with Ω thereby indicating that the coherence between $|1\rangle$ and $|2\rangle$ also grows and finally saturates at a large value of Ω .

In case of a multi-level system, the above discussion can be extended and one can arrive at the generalised form of ρ_{ji} as a function of δ_{ji} . Here, corresponding auto-correlations of the density matrix elements survive also between $|1\rangle$ and $|3\rangle$, $|1\rangle$ and $|5\rangle$ and, $|1\rangle$ and $|7\rangle$ and so on, via multi-photon resonant couplings (see Fig. 1b for nomenclature of states). For clarity, we have named the excited and ground states participating in the long-range coupling as even and odd states, respectively. These additional autocorrelations are the observed higher spin harmonics in the measured Raman driven Spin Noise signal discussed in the next section. For the case of multi-level Raman coupling, the relative transition rate (strength of $(2n - 1)^{\text{th}}$ state to that of state $|1\rangle$) is proportional to $(\frac{\Omega^2}{\gamma\gamma'})^{2n-2}$. The details of the mathematical formalism can be found in²⁰. We note that this ratio depends mainly on three important parameters, which are γ , γ' and Ω . This ratio increases with Rabi frequency Ω (intensity of the Raman beam) and shows a fall if γ and γ' rise, i.e. when the system has faster decay rates.

Results and discussion

In general, atomic systems are more complex due to the presence of multi-level atomic structure. Hence, the simple 3-level models like the Lambda or V-type system do not completely capture the dynamics of the system. Instead, they can be better described by a more intricate configuration like a cascaded-lambda-system (CLS)^{26,27} or ladder-type system²⁸. In this section, we present our experimental observations of relative harmonic strengths as we systematically vary key parameters, including atomic density, Raman drive strength, probe beam intensity, and the applied magnetic field.

Multi-level coupling

Building on our previous demonstration that a two-photon drive yields a 10^6 amplification of the intrinsic SNS signal^{24,25,29}, we mapped the driven spin-noise spectrum. In the presence of a magnetic field, the multi-level structure of the ground and excited state hyperfine manifold ($^2S_{1/2}$, $F = 3$ and $^2P_{3/2}$, $F' = 4$) gets further split. In this case, the ground-state manifold F expand the accessible transitions beyond a single three-level Λ to a CLS (see schematic in Fig. 1b). Figure 2 shows the measured RDSNS spectrum. Instead of a single resonant peak at the drive frequency, the spectrum contains five distinct peaks located at ν_L , $2\nu_L$, $3\nu_L$, $4\nu_L$, and $5\nu_L$. The peaks are separated by exactly $\nu_L = 2.38$ MHz and form a clear frequency comb in the spin-noise signal. These additional spectral lines are consistent with coherent coupling across multiple Zeeman sublevels and with multi-photon mixing among cascaded Raman pathways. In other words, the two-beam drive opens multiple driven Raman channels that produce discrete resonances at integer multiples of ν_L .

These lines arise from driven transitions among Zeeman sub-levels of the multi-level ground-state manifold. When the two Raman beams synchronously drive the spins, atoms are coherently transferred between the nearest-neighbour magnetic sub-levels at the frequency difference δ_{12} , producing the primary RDSNS peak. In addition to the nearest neighbour level transitions, the atomic population may be driven between the next-nearest (nn) and next-next-nearest (nnn) magnetic sublevels with a stronger drive, opening additional Raman pathways. Each of these higher-order transitions produces a spin-noise resonance at an integer multiple of δ_{12}

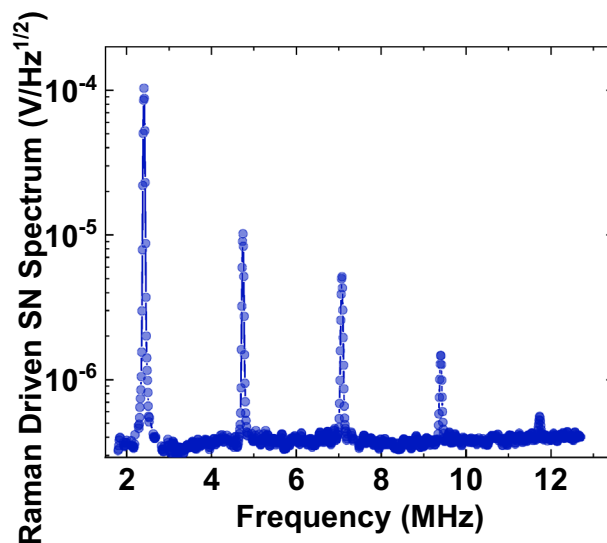


Fig. 2. Shows the observation of multiple peaks in the Raman driven spin noise spectrum. The probe beam power was 0.64 mW, and the Raman beam power was 4.42 mW. The cell temperature was maintained at 95°C . The Larmor frequency was 2.38 MHz.

(e.g., $2\delta_{12}$, $3\delta_{12}$, ...), which appears as the observed harmonic comb. The peak height becomes strongest at ν_L when the driving frequency δ_{12} coincides with the Larmor frequency (ν_L). This has been shown and discussed in detail in “[Detection sensitivity of external magnetic field](#)” section where the driving frequency has been swept across resonance. This peak represents the atomic ensemble’s resonant spin precession in response to an external magnetic field. Due to multi-photon coupling, the selection rule gets altered and hence makes such observations possible. We do, however, observe a succession of weaker peaks at integer multiples of the Larmor frequency. The atomic population that can be coherently driven across increasingly widely separated magnetic (Zeeman) sublevels is progressively reduced. This reduction in the participating population is consistent with the systematic drop in the harmonic intensity. A simple, theory-consistent prediction emerges and matches our data. The relative intensity of the n^{th} harmonic (defined as $R_n = I_n/I_1$ where I_n is the peak strength of the n^{th} peak) increases with the Rabi frequency Ω , because stronger driving enhances the multi-photon coupling amplitudes that generate coherences between more widely separated Zeeman sublevels. Consequently, the harmonic amplitudes grow with drive strength, and are eventually expected to saturate at very large Ω as it no longer transfers additional population coherently.

As seen in Fig. 3, we specifically assess the peak strength ratios between the first, second, third and fourth harmonics with respect to that of the fundamental peak. As the intensity of the Raman beams increases, the Rabi frequency also increases. This results in stronger two- and multi-photon coupling. The observed hierarchy of harmonic strengths: the first harmonic is stronger than the second, the second stronger than the third, and so on, because (i) progressively fewer atoms can be coherently driven into the more widely separated sublevels required for higher-order transitions, and (ii) each additional order is increasingly sensitive to decoherence and imperfect coupling. In short, the measured increase of R_n with drive and the systematic falloff of R_n with harmonic order are direct, qualitative consequences of the multi-photon Raman coupling amplitudes and the competition between Ω and the decoherence rates γ , γ' . In practice, the parameters employed keep the system in the linear-response regime, well separated from the onset of saturation effects. On a related note, reference³⁰ presented a theoretical description of the higher harmonics in spin noise as a function of AC drive strength. Our RDSNS higher harmonics with the Raman drive strength have a qualitative agreement with those results.

Another important feature to note here is that the peak ratios only begin to grow after a critical threshold Raman intensity is reached. In this example, the R_2 , R_3 , R_4 and R_5 starts to only rise after around $8.0 \times 10^{-3} \Omega/\Gamma$, $8.5 \times 10^{-3} \Omega/\Gamma$, $9.5 \times 10^{-3} \Omega/\Gamma$ and $12.0 \times 10^{-3} \Omega/\Gamma$ respectively. The threshold is seen to increase with the order of harmonics. This is due to the requirement for greater intensity to achieve a similar amount of efficient population transfer across higher cycles for long-range couplings. One can optimise the signal-to-noise ratio by fine-tuning experimental parameters and provide insight into the dynamics of coherent light-matter interactions. Knowing how harmonic peak strength ratios alter with the driving strength provides important insights into the underlying quantum processes like stimulated Raman scattering and multi-photon transitions that shape the observable spectra.

Density dependence of harmonic formation

In this section, we discuss how the atomic density influences the shaping of the harmonic structure of the spin noise signal. Since the density determines the effective number of atoms being probed, variations in density strongly influence both the amplitude and spectral features of the signal. It is well established that the overall spin noise amplitude increases with atomic density, which can be controlled by the temperature of the vapour

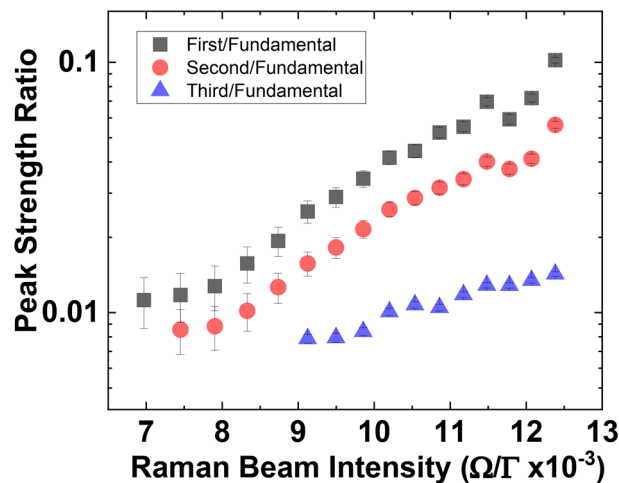


Fig. 3. Dependence of harmonic peak strength ratios on the total Raman beam intensity at a cell temperature of 95°C , corresponding to an atomic density of 4.5×10^{12} atoms/cc. The total Raman beam power was varied from 0.42 to 4.42 mW. The ratios of the first (black squares), second (red circles) and third (blue triangles) harmonics to the fundamental peak are shown as a function of Raman beam intensity.

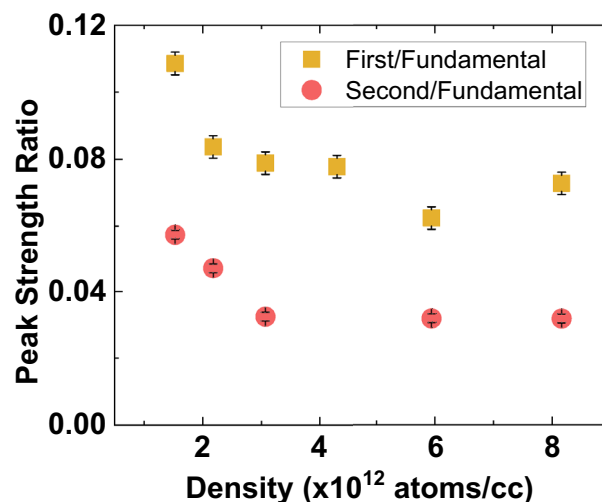


Fig. 4. Shows the dependence of the peak strength on the density of atoms. The density inside the vapour cell was varied by changing the temperature from 80°C to 105°C , and the peak strength ratio of the first harmonic (yellow squares) and second harmonic (red circles) to the fundamental peak was measured.

cell, since a larger ensemble contributes to the collective fluctuations^{3,4}. However, a closer look at the harmonic spectrum reveals a more nuanced and somewhat counterintuitive behaviour. Figure 4 shows the ratios of the first and second harmonic peak strengths relative to the fundamental. Both ratios decrease as the density increases, indicating a redistribution of driving intensity across the harmonic orders, which leads to further dephasing of the spin ensemble. We have shown earlier²⁴, the fundamental peak continues to grow with density; in contrast, the higher-order harmonics systematically weaken. This behaviour is closely linked to the increasing role of collisional processes, both atom–atom and atom–wall collisions, which disrupt coherent spin precession and cause dephasing. In theoretical terms, the harmonic ratios R_n are expected to decrease with increasing decoherence rates γ and γ' , as faster dephasing limits the population that can participate coherently in higher-order processes. Collisions effectively push atoms out of the coherent spin cycle, reducing their ability to remain synchronised with the driving field. This dephasing manifests as an exponential suppression of harmonic peak ratios with rising density. The interplay between enhanced driving of the fundamental and increased collisional dephasing of the harmonics creates a characteristic density-dependent pattern. This sheds light on the underlying physics of spin noise spectroscopy (SNS) in dense atomic systems, where driven and undriven cases exhibit strikingly different trends. Understanding this balance is also crucial for experimental optimisation: by carefully tuning cell temperature, probe intensity, and atomic density, one can either enhance or suppress specific harmonics depending on the desired application.

Beyond optimising sensitivity in quantum sensing and metrology, this behaviour also suggests practical applications. Since the harmonic ratios vary systematically with density, a calibrated vapour cell could be employed as a *density sensor*, providing a direct and noninvasive diagnostic tool. In this work, the atomic density was varied over a relatively narrow range of $1 - 8 \times 10^{12}$ atoms/cc, which is sufficient for observing the key features reported here. Extending the platform to higher densities could enable the study of stronger collisional effects.

Probe power dependence

In Spin Noise Spectroscopy, the signal level increases with increasing probe beam intensity. Increasing the intensity of the probe beam increases the atom-photon interaction strength. This gives rise to greater collective spin fluctuations. Spin fluctuations (Spin Noise) of the atoms cause polarisation fluctuations of the probe beam. These fluctuations are proportional to the spin fluctuations. Increased probe intensity amplifies these polarisation fluctuations, making them detectable. At very low probe intensities, the spin fluctuation signals are so small that they are masked by the inherent noise of the system. At higher probe power, the same fluctuations become more visible as they go above the noise floor.

Higher probe intensities increase the collective spin precession's magnitude and strengthen the spin noise signal across all harmonic components. This phenomenon is well described by the experimental plot in Fig. 5, where the harmonic strengths are studied over a range of probe power. Raising the intensity of the probe beam can lead to saturation effects beyond a certain intensity, where additional power no longer results in an increase in signal intensity. Additionally, very high probe intensities would have unintended consequences, such as atomic collisions and dephasing, which would negate the benefits of the increased intensity. Optimising the harmonic content of the spin noise signal is possible by carefully tuning the probe beam intensity. In conclusion, our findings show that increasing the probe beam intensities increases the higher harmonic peaks in relation to the fundamental.

Magnetic field dependence

The spin noise signal's harmonic structure is closely tied to the changes in the external magnetic field. This is evident from our observation, illustrated by Fig. 6. Here, we changed the magnetic field from 5 G to 10 G, and we see that the strength of the higher harmonics reduces as the field is increased. This pattern arises from the fundamental nature of the spin noise signal, which is generated by atomic population fluctuations among the two ground-state hyperfine levels' different magnetic sublevels, or m_F states. The energy difference between these states is governed by the Zeeman effect. This effect causes the energy splitting between the m_F states to become more pronounced as the magnetic field is increased. This has a direct impact on the interaction of the Raman driving beams with the atoms. The Raman fields couple several m_F states when the field is weak, allowing strong spin noise signals on all harmonics and coherent population oscillation. This results in long-range coherence between all the coupled Zeeman states. As the intensity grows, the Raman beams, which were initially tuned to couple transitions among m_F states, become increasingly detuned from the higher-order transitions. The weaker coupling between the Raman fields and multiple m_F states is the reason attributed to the decrease in higher harmonic peak strength. Consequently, effective population transfer between the coupled states is reduced. This leads to reduced strength of the higher-order harmonics that correspond to transitions involving these higher m_F states with increasing magnetic field strength.

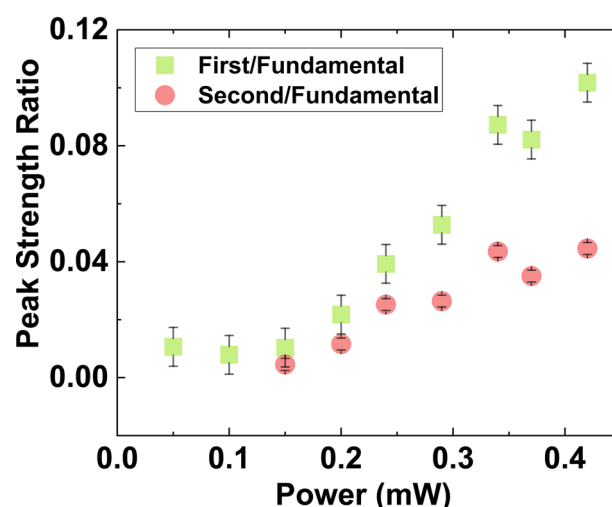


Fig. 5. Illustrates dependence of the harmonic peak strength ratio on probe beam power. Increasing probe intensity enhances atom–photon interaction, strengthening both first and higher harmonic peaks. At low power, the signal remains buried in noise, while at higher powers, saturation and dephasing limit further growth. The optical power indicated in the plot corresponds to the power measured in one arm of the balanced detector, which is half of the total optical power.

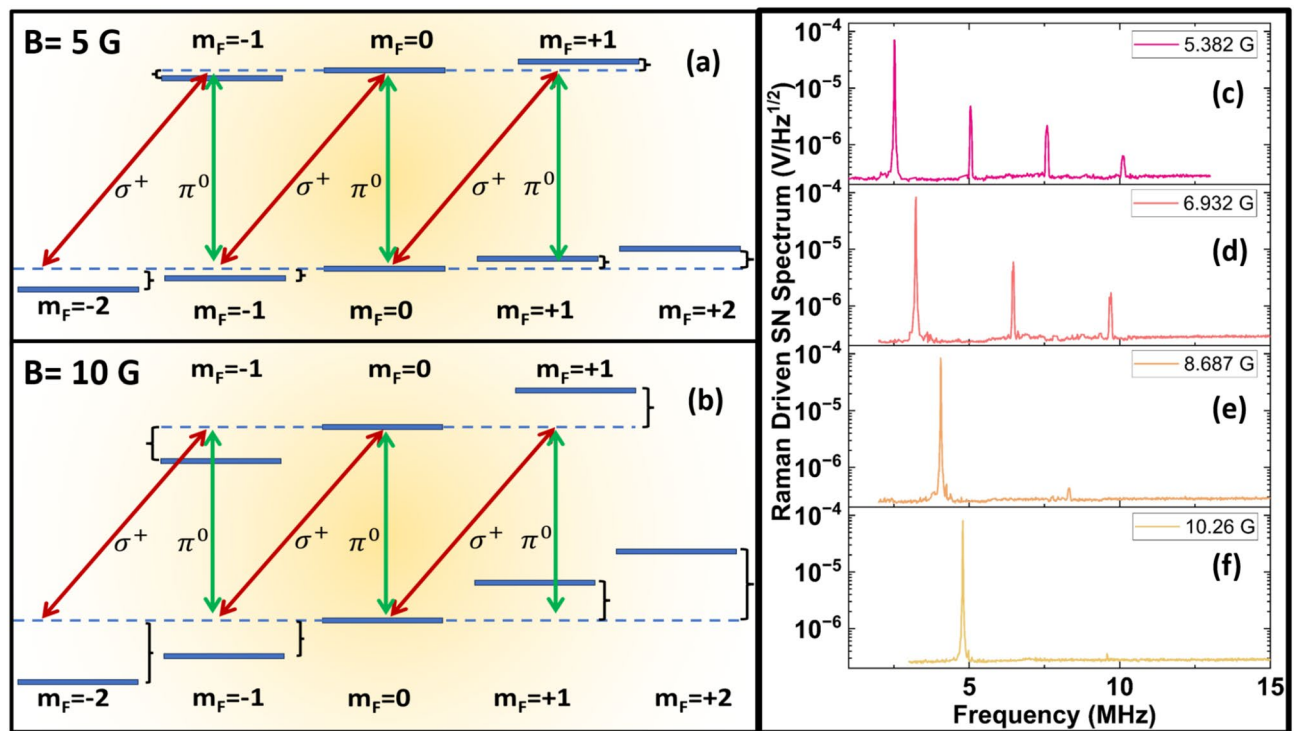


Fig. 6. Shows the study of the effect of external magnetic field on the harmonic structure of the spin noise peaks. Schematic of Raman coupling between Zeeman sublevels (m_F states) of the ground-state hyperfine manifold (a) at low field and (b) at high field. Red arrows indicate σ^+ transitions and green arrows indicate π^0 transitions, forming cascaded multi- Λ pathways. (c–f) Raman driven spin noise spectra recorded at different magnetic field strengths (5.38 G, 6.93 G, 8.69 G, and 10.26 G). The fundamental Larmor peak is visible in all cases, while higher-order harmonics weaken progressively with increasing magnetic field due to reduced resonance overlap and detuning of Raman coupling from higher-order transitions.

Additionally, a higher magnetic field causes the atomic spins to Larmor precess more quickly, which can reduce the coherence time of the atomic ensemble. Phase mismatches and dephasing effects are introduced in this domain due to the various frequencies at which atoms in different m_F states precess. As the coherent population oscillations required to produce large harmonic peaks become harder to maintain, these effects can further limit the contributions from higher-order harmonics. In precision metrology and quantum sensing applications, where control over the magnetic environment can be utilised to improve measurement resolution and accuracy, the reliance of the harmonic structure on the magnetic field also has wider ramifications. One way to increase measurement sensitivity and target particular quantum states with the signal could be to carefully adjust the Raman beam detunings and magnetic field. The harmonic structure of the spin noise signal may be successfully controlled by adjusting the magnetic field, which also affects the coupling between the Raman beams and the atomic transitions. Depending on the intended measurement results, this control can be used in experimental situations to deliberately amplify or suppress certain harmonic components. Stronger fields can be helpful for exploring certain m_F transitions or isolating the traditional spin noise signal, but lower fields could be desirable when a broad harmonic response is required. The reduction in the number of harmonics at high magnetic field imposes a practical limitation on the dynamic range of the scheme for magnetic-field sensing.

Detection sensitivity of external magnetic field

In^{24,25}, it was shown that when the frequency difference between the two Raman beams is varied, δ_{12} , a prominent peak appears at δ_{12} . The peak strength rises as δ_{12} approaches closer to the Larmor frequency ν_L and reaches a maximum exactly at ν_L . This is a characteristic of the Raman resonance situation, in which the Raman drive frequency matches the energy splitting between the two ground-state hyperfine levels. The strongest signal results from the most effective population oscillation between the spin states. Interestingly, we see a similar tendency in the higher harmonics of the spin noise signal, indicating that this resonance characteristic is not limited to the fundamental peak. At twice and thrice the Larmor frequency, the strength of the first harmonic and the second harmonic reaches its maximum. We illustrate this pattern in Fig. 7, where we find that as δ_{12} approaches multiples of the Larmor frequency, the higher harmonic peaks similarly follow this resonance-like trend, much like the fundamental peak. The fact that the higher harmonic peaks only start to show up when δ_{12} approaches very near to the Larmor frequency is noteworthy. This trend implies that relatively small variations in the frequency difference between the Raman beams and the Larmor frequency have a considerably greater sensitivity on the higher harmonics than on the fundamental peak. The higher harmonics are muted until the

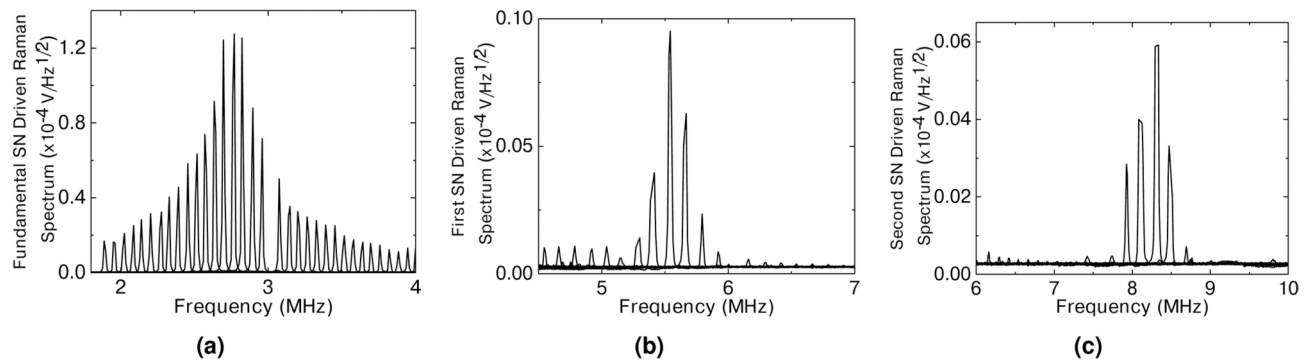


Fig. 7. Illustrates the dependence of drive frequency on the Raman Driven Spin Noise Signals. The temperature is maintained at 95°C . The total Raman beam intensity was kept at 3.75 mW. The probe beam power was 0.7 mW. Each black peak corresponds to a particular Raman driving frequency. (a) Shows the fundamental peaks when the driving frequency (δ_{12}) is scanned across the Larmor frequency (ν_L). Strongest peak appears when $\delta_{12} = \nu_L$. (b) Shows the occurrence of the first harmonic peaks with the strongest peak appearing at $2\nu_L$. (c) Shows the observation of the second harmonic peaks with the strongest peak appearing at $3\nu_L$.

Raman frequency difference is exactly or extremely closely tuned to ν_L , although the fundamental peak still emerges even when δ_{12} is significantly detuned from ν_L . Due to their increased sensitivity, the higher harmonics can be used as an effective probe for detecting small changes in magnetic fields. This is because even slight changes in the magnetic environment can cause the resonance state to change, which can have a big impact on how visible these harmonics are. The observed linewidths of the Raman driven peaks are constrained by relative AOM frequency jitter (5 kHz). A phase-locked, frequency-stabilised Raman pair would mitigate this limitation which is a refinement we reserve for future work. A slight asymmetry is observed in the spectral envelope around the harmonic peaks. This can be attributed to magnetic field inhomogeneity across the interaction region, which results in a spread of Larmor frequencies and hence an asymmetric broadening of the spectrum. Importantly, the peak positions remain fixed at integer multiples of ν_L , confirming their origin in higher-order Raman driven spin noise peaks.

Conclusion and outlook

We have shown the observation of multiple peaks in the spin noise spectrum and explored its occurrence by systematically changing experimental parameters, such as the Raman driving frequency, magnetic field, probe intensity, Raman beam intensity, and atomic density. The phenomenon of multiple, equally spaced peaks is explained by long-range multi-level coupling in the ground-state Zeeman manifold, induced by Raman fields with particular polarisations. These peaks are highly sensitive to small fluctuations in the magnetic field, making them a strong candidate for a quantum sensor. These findings are consistent with earlier work demonstrating SNS can measure tiny magnetic field fluctuations and spin interactions with high spectral resolution^{31,32}.

The recorded progression of harmonics represents a driven *spin-noise comb*, somewhat analogous to an optical frequency comb³³. Consequently, adding an external cavity with high finesse would sustain higher modes in such a driven spin noise system. In addition, the inherent shot-noise limitation of SNS can be overcome by using a phase-squeezed probe light^{34,35}, which reduces the noise floor and increases the visibility of the higher harmonics.

Data availability

The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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Author contributions

S.C. conceived the experiment and secured funding, S.M. conducted the experiment and analysed the results. All authors wrote and reviewed the manuscript.

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Declarations

Competing interests

The authors declare no competing interests.

Additional information

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