

Quantum Memory Enhanced Multipoint Correlation Spectroscopy for Statistically Polarized NMR

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Nuclear magnetic resonance spectroscopy with solid-state spin sensors is a promising pathway for the detection of nuclear spins at the micro- and nanoscale. Although many nanoscale experiments rely on a single sensor spin for the detection of the signal, leveraging spin ensembles can enhance sensitivity, particularly in cases in which the signal merely originates from statistically polarized nuclear spins. In this Letter, we introduce multipoint correlation spectroscopy, which combines the advantages of two well-established methods—correlation spectroscopy and quantum heterodyne detection—to enable temporally efficient measurements of statistically polarized samples at the nanoscale with spin ensembles. We present a theoretical framework for this approach and demonstrate an experimental proof of concept with a nitrogen vacancy center in diamond. We achieve single hertz uncertainty in the estimated signal frequency, highlighting the potential applications of the technique for nanoscale nuclear magnetic resonance.

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Nuclear magnetic resonance (NMR) has been widely used for multiple applications in physics, chemistry, biology, and medicine for the past several decades. However, detection of a signal typically requires significant thermal polarization of a nuclear spin sample, which generally exceeds several μm^3 [1–3]. The thermal polarization increases when stronger bias magnetic fields are applied, rendering high magnetic fields common for NMR experiments. Solid state spin sensors, such as the negatively charged nitrogen-vacancy (NV) center in diamond, allow for a significant reduction of the sensing volume to a few nm^3 [4–6]. Then, statistical polarization becomes dominant, especially at low magnetic fields [7,8]. In this regime, the polarization follows Poissonian random statistics $\propto N_{\text{nuc}}^{-1/2}$ and is dependent only on the number of nuclear spins N_{nuc} in the sensing volume and no longer on the bias magnetic field [4,5]. This enables applications such as single molecule [9] or spin [10] spectroscopy and the determination of nanoscale structures at ambient conditions [11,12].

Several measurement protocols have been established to detect nano- and microscale NMR signals with NV centers,

most prominently correlation spectroscopy (CS) [13–17], quantum heterodyne detection (QDyne) [8,18,19], and CASR [20–22]. CS correlates the phases ϕ_0, ϕ_k acquired at times T_0, T_k , respectively, by a quantum sensor during two sensing periods, as shown in Fig. 1(a). The phase ϕ_0 is stored in the populations of the quantum sensor [13–15] or an ancillary memory qubit [16,17,23]. By sweeping k , the autocorrelation of the accumulated phases ϕ_0 and ϕ_k is detected. The frequency resolution of this method is limited by the spin lifetime of the NV center [13–15] or that of the ancillary memory qubit [16,17,23], where the latter can be several orders of magnitude longer [16], leading to improved resolution. A major disadvantage is the typically long idle time between the two phase acquisition periods when no data are acquired. Furthermore, multiple measurements have to be performed, where the idle waiting time is varied, in order to obtain the phases correlation function.

QDyne mitigates this problem by repeating the measurement at a constant rate, determined by the time needed for initialization, detection, and readout [18,19], as visualized in Fig. 1(b). This significantly reduces the temporal overhead of the measurement, resulting in efficient signal acquisition. The correlations are established indirectly and are retrieved in postprocessing. However, QDyne is limited by the low readout efficiency for single NV centers due to photon shot noise [8]. The experiments then require a longer measurement time to reach a sufficient signal-to-noise ratio (SNR), limiting sensitivity.

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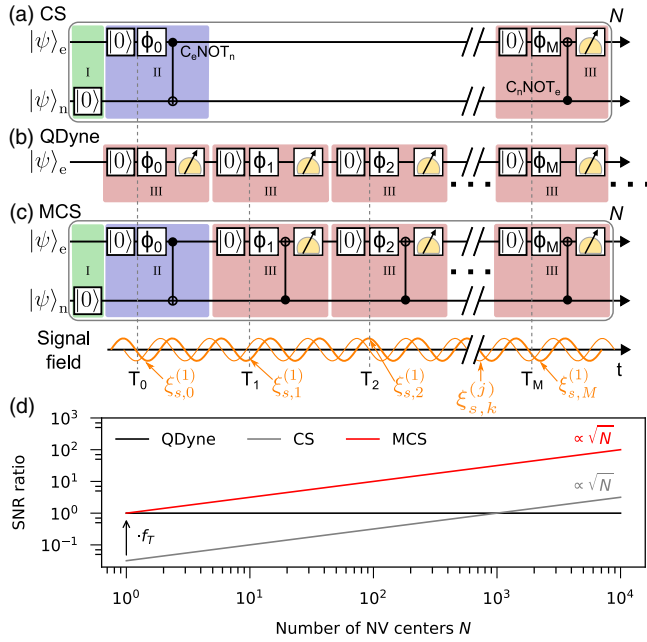


FIG. 1. (a) CS measurement scheme with ancilla based memory. (I) The memory spin n is initialized before (II) acquiring an initial phase $\phi_0^{(j)}$ with the sensor spin e , where (j) denotes the j th repetition of the experiment or the j th NV in an ensemble, $j = 1, \dots, N$ (j omitted for simplicity in the figure) and mapping it onto the memory spin by using a $C_e \text{NOT}_n$ gate. In a subsequent phase measurement (III) the phase $\phi_k^{(j)}$ ($k \in \{1, \dots, M\}$) is acquired, which is correlated to the initial phase by performing a $C_n \text{NOT}_e$ gate. The phases $\phi_k^{(j)}$ depend on the signal phase $\xi_{s,k}^{(j)}$ at the beginning of the respective phase acquisition. (b) QDyne measurement scheme. Only a sensor spin e is used to acquire the phases ϕ_k at a constant rate. Commonly, the protocol is performed for more than $M + 1$ phase acquisitions in order to improve the SNR. (c) MCS scheme. Similar to CS, a sensor and memory spin are used to establish correlations between an initial phase ϕ_0 and the measurement phases ϕ_k by storing and retrieving ϕ_0 on the memory spin. $M + 1$ phase acquisitions are performed during one sequence, which can be repeated N times. (d) Comparison of the SNR of the three protocols vs the number N of NV centers in an ensemble, where photon shot noise is assumed to be the main noise source. SNR of MCS and CS (QDyne) protocols are normalized to the one of MCS (QDyne) with a single NV center. $\text{SNR}_{\text{QDyne}}$ does not change with N while SNR of both CS and MCS $\propto \sqrt{N}$. MCS is favorable by factor $f_T \propto \sqrt{M}$ to CS as its data acquisition lacks long waiting times.

Using multiple NV centers in an ensemble simultaneously can significantly improve sensitivity [24]. However, the SNR of QDyne in nanoscale NMR does not improve with a larger number of NV centers. While the photon shot noise decreases, the detected signal also drops with statistical polarization. Specifically, the photoluminescence signal from the j th NV center is linearly dependent on its acquired phase $\phi_k^{(j)}$, which varies for each NV center as

they have different sensing volumes. As all NV centers are read out simultaneously, averaging is performed over all these phases. This leads to an effective increase in the sensing volume, resulting in a reduction in the signal detected from statistical polarization [4,5]. As a result, there is typically no net benefit when using QDyne with an NV ensemble (see Supplemental Material [25]). By contrast, CS, with its direct detection of phase correlations, detects a signal, which is proportional to the average phase variance for each NV center. Thus, the effective sensing volume does not increase, making CS applicable to NV ensembles [7].

In this Letter, we introduce multipoint correlation spectroscopy (MCS), which combines the advantages of QDyne and CS, i.e., the high data acquisition rate of the former and CS's improvement of sensitivity with multiple NV centers without the loss of statistical polarization signal. Specifically, MCS detects the correlation of an initial phase ϕ_0 with many subsequent phases ϕ_k , acquired in a QDyne-type fashion. By storing the information about ϕ_0 in a memory qubit, the following phases ϕ_k can be correlated with ϕ_0 . The protocol is limited by the lifetime of the memory qubit, which can reach up to a second for the intrinsic nitrogen nuclear spin [26], allowing for single-hertz resolution.

Theory—The theoretical description of MCS is general as it is applicable to any pair of a sensor (e) and a memory (n) qubit for advanced quantum sensing. Figure 1 shows a comparison of CS, QDyne, and MCS. In our particular experiment, we use a negatively charged NV center in diamond with its electron spin—the sensor qubit, and the nuclear spin of nitrogen—the memory qubit. To match its description, the states are labeled as $|m_s\rangle_e |m_1\rangle_n$, corresponding to $\{|0\rangle_e |0\rangle_n, |0\rangle_e |1\rangle_n, |1\rangle_e |0\rangle_n, |1\rangle_e |1\rangle_n\}$, (see Fig. 2 and [25]). The system is initialized in state $|0\rangle_e |0\rangle_n$ [step I in Fig. 1(c)]. For simplicity of presentation, initialization is assumed as ideal. Imperfect initialization would lead to reduced contrast [25].

MCS then includes an initial phase acquisition (step II), where a signal-dependent phase ϕ_0 is accumulated and subsequently stored as a population difference in the ^{14}N nuclear spin memory qubit. We apply a $\pi/2$ pulse on the sensor spin to create a coherent superposition state. Then we use dynamical decoupling (DD) with the XY8 [27] sequence to increase the coherence time and allow for the detection of weak oscillating signals [28]. Robust DD sequences [27,29–32] allow for phase acquisition with NV ensembles where frequency and amplitude inhomogeneities might be present [25]. We apply DD on resonance, i.e., the time between the centers of the π pulses is $\tau \approx 1/(2\nu_s)$ [28], where $\nu_s = \omega_s/(2\pi)$ is the frequency of the signal and ω_s its angular frequency. In the case of nano-NMR, based on statistical polarization, ν_s is the expected Larmor frequency of the nuclear spins ensemble [7]. The accumulated phase is [28]

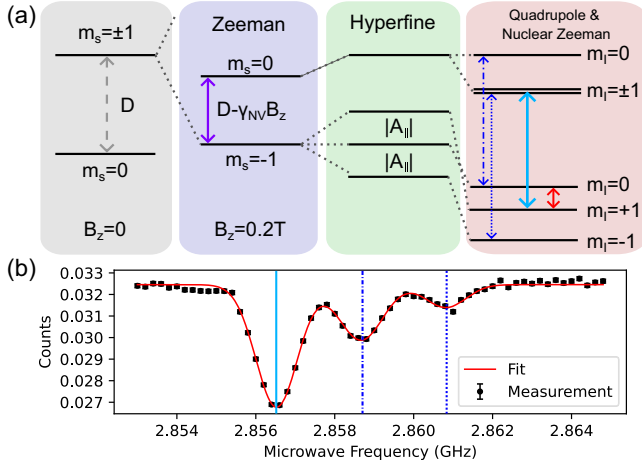


FIG. 2. (a) Level scheme of an NV center. Electronic spin levels $m_s = 0, \pm 1$ are split by the zero field splitting D and Zeeman interaction $\pm \gamma_{\text{NV}} B_z$, creating nondegenerate spin levels. Only the subsystem $m_s = 0, -1$ is considered. Hyperfine A , quadrupolar P , and nuclear Zeeman $\gamma_{\text{N}} B_z$ interaction with the intrinsic ^{14}N nuclear spin splits the electron spin states into three additional sublevels $m_1 = 0, \pm 1$. (b) Pulsed ODMR measurement reveals the hyperfine structure of the NV center with intrinsic ^{14}N for the $m_s = 0 \leftrightarrow m_s = -1$ transition. 89.1(18)% of the nuclear spin population is in the $m_1 = 0, +1$ states. The dashed lines show the corresponding microwave transitions in (a), which are separated by the hyperfine coupling of approximately 2.1 MHz. The protocol was demonstrated, using the red (light blue) transition for RF (selective and strong MW) pulses.

$$\phi_0^{(j)} \approx \frac{2}{\pi} g_{s,0}^{(j)} t_{\text{DD}} \cos(\xi_{s,0}^{(j)}) = \frac{2}{\pi} \gamma_{\text{NV}} B_{s,0}^{(j)} t_{\text{DD}} \cos(\xi_{s,0}^{(j)}), \quad (1)$$

where $g_{s,0}^{(j)}$ is the signal amplitude, which is proportional to the signal magnetic field envelope $B_{s,0}^{(j)}$, t_{DD} is the XY8 duration, and $\xi_{s,0}^{(j)}$ is the signal phase at time T_0 when detection starts. The superscript $j \in \{1, \dots, N\}$ denotes the respective value for the j th sensor in an ensemble or the j th experimental run when the measurement is performed on a single spin pair. The phase information $\phi_0^{(j)}$ is then transferred to the populations of the sensor spin states by a $\pi/2$ Y pulse and is subsequently stored in the population of the memory qubit by a π pulse on the memory qubit, conditioned on the spin state of the sensor, which we label a $C_n \text{ NOT}_e$ gate.

In step III, we perform a series of subsequent phase measurements. In each experiment, the sensor is prepared in the state $|0\rangle_e$, ideally without affecting the memory qubit. Then, we repeat the previous step and the sensor accumulates a phase

$$\phi_k^{(j)} \approx \frac{2}{\pi} g_{s,k}^{(j)} t_{\text{DD}} \cos(\xi_{s,k}^{(j)}), \quad (2)$$

where $g_{s,k}^{(j)} = \gamma_{\text{NV}} B_{s,k}^{(j)}$ is the signal amplitude during the k th period. The signal has the phase $\xi_{s,k}^{(j)} = \xi_{s,0}^{(j)} + \omega_s \Delta T_k$ at the beginning of k th detection with $\Delta T_k = T_k - T_0$. Information about the initial period phase $\phi_0^{(j)}$ is retrieved after every phase acquisition with a π pulse on the sensor spin, conditioned on the memory spin state, which we label $C_n \text{ NOT}_e$ gate. The population of the sensor spin state $|0\rangle_e$ and the detected signal depends on the correlation between $\phi_0^{(j)}$ and $\phi_k^{(j)}$ [25]:

$$\begin{aligned} S_k &\approx \eta \left(1 + \frac{c}{2} \langle \phi_0^{(j)} \phi_k^{(j)} \rangle \right) \\ &= \eta \left(1 + \frac{c}{2} \frac{4}{\pi^2} \gamma_{\text{NV}}^2 B_{\text{rms}}^2 t_{\text{DD}}^2 \cos(\omega_s \Delta T_k) \right), \end{aligned} \quad (3)$$

where $\eta = (\eta_0 + \eta_1)/2$ is the average signal, typically the number of photons, with η_0 (η_1) the expected signal when the sensor qubit is in state $|0\rangle$ ($|-1\rangle$), and $c = (\eta_0 - \eta_1)/\eta$ is the relative experimental contrast. The phase $\phi_k^{(j)}$ is assumed to be small, so that $\sin(\phi_k^{(j)}) \approx \phi_k^{(j)}$ for the first equality. The averaging $\langle \cdot \rangle$ is done on the $j = 1, \dots, N$ different sensors in the ensemble or over the N repetitions of the experiment on a single spin pair. The quantity $B_{\text{rms}}^2 = B_s^2/2$ for a classical signal with a constant amplitude $B_{s,0}^{(j)} = B_{s,k}^{(j)} = B_s$ for all measurements (or all spin pairs in an ensemble), or B_{rms} is the root mean square of the magnetic field due to statistical polarization, sensed by a sensor spin in an ensemble [7,8,25]. The signal obtained after every k th measurement allows for extraction of the correlation between the accumulated phase during the k th measurement and during the initial measurement. The signal scales as $S_k \sim \eta \sim N$, while the usually dominant photon shot noise $\sim \sqrt{N}$, leading to improved SNR $\sim \sqrt{N}$, as shown in Fig. 1(d). This is a significant advantage over Qdyne with spin ensembles, where the signal from the k th measurement depends on $\langle \phi_k^{(j)} \rangle \sim B_{\text{rms}}/\sqrt{N}$ for $j = 1, \dots, N$, meaning that SNR_{Qdyne} does not improve for ensembles with N spins compared to the single spin case [25]. Although the signal oscillates with an angular frequency ω_s , measurements at times T_k typically detect an undersampled frequency $\omega_u = (2\pi)f_u$, related to ω_s [7,8,18], which can be extracted by performing a Fourier transform. As the detected signal is related to the autocorrelation function of the sensed magnetic field, the Fourier transform produces its power spectral density [25]. The line width is $\sim 1/T_{\text{total}} \approx 1/\Delta T_M$, where M is the total number of phase acquisitions after the initial phase accumulation. The total measurement time T_{total} is limited by the lifetime $T_{1,\text{nuc}}$ of the memory spin qubit, considering that it could be affected by the initialization of the sensor spin [25]. The key advantage of MCS over CS is an improved SNR by factor:

$$f_T = \sqrt{\frac{M}{2} \left(1 + \frac{1 + T/T_{\text{init}}}{1 + MT/T_{\text{init}}} \right)}, \quad (4)$$

where T_{init} is the total duration of steps I and II and $T = T_{k+1} - T_k$ is the duration of one measurement in step III [25]. This improved SNR is caused by reducing measurement idle time between the two phase acquisition periods of CS and replacing it with repeated phase measurements.

Experimental demonstration—We demonstrate the applicability of the protocol with a single NV center with an intrinsic ^{14}N nuclear spin. Its level structure at a magnetic field of 2043.763(6) G is shown in Fig. 2(a) with only the subsystem $m_s = 0, -1$ being used as the sensing qubit. Measurements are carried out using a home-built confocal microscope, using the Qudi measurement suite [33]. The MCS pulse sequence for an NV center is shown in Fig. 3(a). First, we perform optical pumping with a green laser pulse to initialize the electron spin of the NV center in $|0\rangle_e$.

Using weak microwave (MW) pulses, the hyperfine structure of the nitrogen nuclear spin becomes visible in an optically detected magnetic resonance (ODMR) spectrum, as shown in Fig. 2(b). This spectrum can be used to determine the degree of polarization of the nitrogen nuclear spin by comparing the area under the three Gaussians

with which the data is fit. The populations of nuclear spin states after long laser irradiation are thus $P_{|0\rangle_e|+1\rangle_n} = 0.606$, $P_{|0\rangle_e|0\rangle_n} = 0.285$, and $P_{|0\rangle_e|-1\rangle_n} = 0.109$ with the corresponding drops in fluorescence denoted from left to right [25]. The goal is to prepare the system in $|0\rangle_e|0\rangle_n$. A selective first MW (subsequent radiofrequency (RF)) π pulse is applied on the transition $|0\rangle_e|+1\rangle_n \leftrightarrow |-1\rangle_e|+1\rangle_n$ ($|-1\rangle_e|+1\rangle_n \leftrightarrow |-1\rangle_e|0\rangle_n$). Then we use optical pumping to reinitialize the electron spin state. In the end, the population of the $m_I = +1$ state [left dip of Fig. 2(b)] is ideally pumped to $m_I = 0$ (central dip) [25]. The procedure can be modified to pump the population of the $m_I = -1$ state to $m_I = 0$, which is not done in this experiment, due to the low population of the former.

Figure 3(b) shows the fluorescence response of the NV center for the MCS protocol, using a test signal of frequency 1 MHz and amplitude 14.28(4) μT . The measured signal oscillates with the undersampled frequency $f_u = 4.2954(13)$ kHz, which we estimate with a precision reaching the single hertz level. The resulting full width at half maximum of the power spectral density, shown in Fig. 3(c), is 59.9(21) Hz. This frequency resolution is directly limited by the nuclear spin lifetime $\tilde{T}_{1,\text{nuc}}$ of the intrinsic nitrogen nuclear spin. During laser irradiation, the intrinsic nuclear spin of the NV center gradually depolarizes [26], limiting its lifetime. A nuclear polarization decay

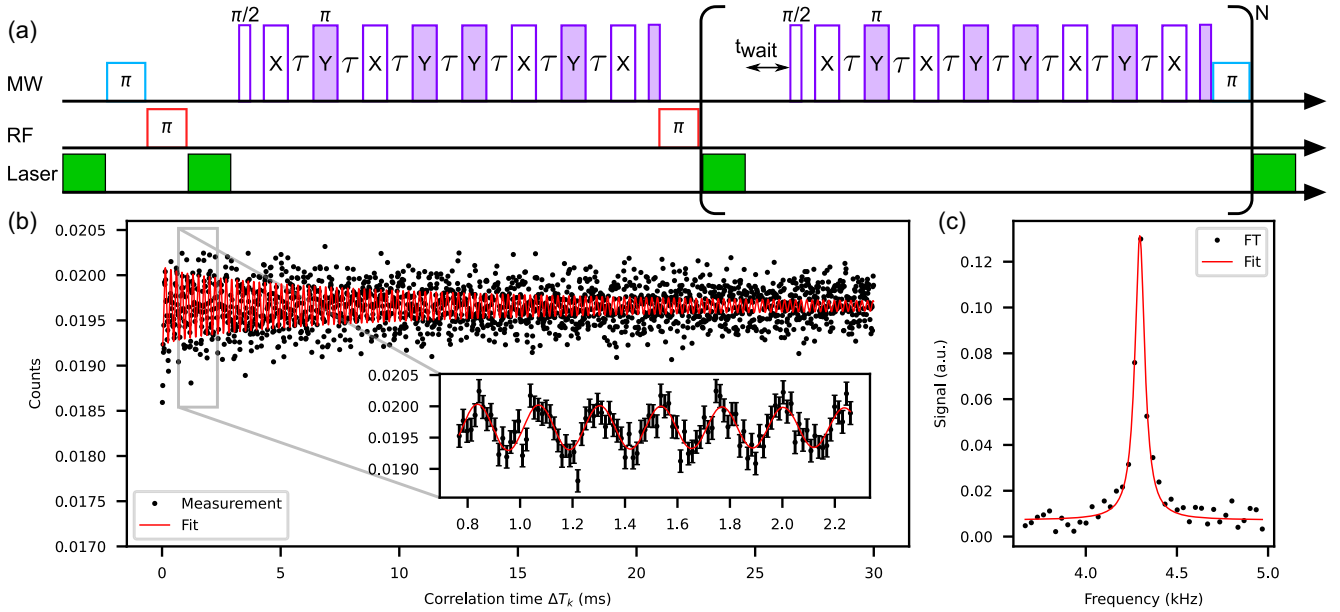


FIG. 3. (a) Experimental scheme of MCS with an NV center in diamond. First, the nitrogen nuclear spin is initialized using selective MW (blue) and RF (red) π pulses. The signal is then probed for the first time, using XY8-1 (purple). The acquired phase is mapped onto the nitrogen nuclear spin by utilizing an RF π -pulse. Subsequently, the phase acquisition is performed $M = 1991$ times, correlating the acquired phase to the phase stored in the population of the nitrogen nuclear spin by applying a selective MW pulse. The sequence is repeated for $N = 597894$ times. (b) Experimentally measured NV center fluorescence response for a 14.28(4) μT signal at 1 MHz when running the sequence in (a). Error bars have been omitted for visibility purposes. Inset: close-up of the data, with error bars corresponding to photon shot noise. (c) Power spectral density of the measured signal. A fit to the signal reveals the detected undersampled frequency $f_u = 4.2954(13)$ kHz with a full width at half maximum of 59.9(2.1) Hz corresponding to a resolution limit due to the nuclear spin lifetime $\tilde{T}_{1,\text{nuc}}$ under laser illumination.

time of $\tilde{T}_{1,\text{nuc}} \approx M_{\text{limit}} T = 15.82$ ms is detected during the measurement, where $T = T_{k+1} - T_k = 15.063$ μs is the time between two subsequent phase measurements and $M_{\text{limit}} = 1050$ is the number of lasers, leading to a $1/e$ decay of the nuclear spin polarization [25]. A line width of $\sim 1/\Delta T_M \approx 33$ Hz would be expected without the effect of laser initialization on the memory spin. In the measurement, the number of acquired phases is $M = 1991$, confirming that the protocol is limited by the shortened nuclear spin lifetime. In order to limit the effect of this decay, a waiting time t_{wait} may be introduced between the M phase acquisitions, reducing the number of laser pulses per unit time and thus prolonging the nuclear $\tilde{T}_{1,\text{nuc}}$ time, which results in a better frequency resolution. This allows for increasing the maximum phase acquisition time ΔT_M of the protocol, resulting in a higher frequency resolution. It can also be improved by increasing the static bias magnetic field B_0 , as the lifetime of the nitrogen nuclear spin under laser illumination depends quadratically on the bias magnetic field [25,26].

The measurement is acquired for 18000 s or $N = 597894$ repetitions, resulting in a sensitivity of 55.64 $\mu\text{T}/\sqrt{\text{Hz}}$. As described in the theory section, this sensitivity can be directly improved by using an NV ensemble to acquire already averaged data within one measurement run. Assuming that all parameters remain constant, using, e.g., 1000 NV centers instead of one, the total measurement time is shortened to ~ 18 s or $N \approx 598$ repetitions, improving the sensitivity to 1.76 $\mu\text{T}/\sqrt{\text{Hz}}$.

Sensitivity can also be optimized by improving the readout signal and contrast, as shown in Eq. (3) [24]. Another limitation is the imperfect initialization of the NV center, which leads to reduced nuclear spin polarization and a limited readout contrast [25]. The complete polarization of the electron [60,61] and nuclear memory spin would thus enhance sensitivity. A second initialization step, transferring the $m_I = -1$ population to $m_I = 0$, could also improve nuclear spin polarization. Using an NV center with a ^{15}N nucleus ($I = 1/2$) would eliminate the need for a second initialization step.

The improved SNR of MCS compared to CS becomes evident when calculating the time required to acquire the data in Fig. 3(b) using CS. A CS measurement would take ~ 999 times longer [25], due to the large measurement idle times and the inability to record the N data points during a single sequence run. This approximately three orders of magnitude improvement in measurement time underscores the temporal advantage that MCS has over CS as the SNR of MCS would be improved by a factor of $f_T \approx \sqrt{999}$ compared to CS, as shown in Fig. 1(d).

Conclusion—We proposed theoretically and demonstrated experimentally multipoint correlation spectroscopy, which uses the intrinsic nitrogen nuclear spin as a memory qubit and combines the benefits of correlation spectroscopy and QDyne protocols for single- and spin-ensemble

measurements of NMR signals. Using QDyne's high data acquisition rate, we significantly reduce the measurement idle time in comparison to correlation spectroscopy. The protocol directly probes the correlation between the accumulated phases in different detection periods, which makes it applicable to both single and ensemble NV center experiments and for detecting signals, based on thermal and statistical polarization. We achieve single hertz uncertainty in the estimated signal frequency in a proof-of-concept experiment in a single NV center, highlighting the broad applications of the technique. Liquid nano-NMR applications are limited by the diffusion of the sample molecules [7,8]. Our protocol does not mitigate this effect. However, several other techniques have been demonstrated that combat diffusion by confining the NMR sample [34–36] or optimizing the NV centers depth, taking into account the trade-off between diffusion broadening and signal strength [62]. Combining these techniques with MCS would allow for significant improvement in the sensitivity of nano-NMR, compared to standard QDyne and correlation spectroscopy.

Note added—Recently, we have become aware of a related independent work [37].

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Data availability—The data that support the findings of this article are openly available [63].

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- [1] C. Munuera-Javaloy, A. Tobalina, and J. Casanova, *Phys. Rev. Lett.* **130**, 133603 (2023).
 - [2] R. D. Allert, K. D. Briegel, and D. B. Bucher, *Chem. Commun.* **58**, 8165 (2022).

- [3] P. Alsina-Bolívar, A. Biteri-Uribarren, C. Munuera-Javaloy, and J. Casanova, *Phys. Rev. Res.* **6**, 043017 (2024).
- [4] H. J. Mamin, M. Kim, M. H. Sherwood, C. T. Rettner, K. Ohno, D. D. Awschalom, and D. Rugar, *Science* **339**, 557 (2013).
- [5] T. Staudacher, F. Shi, S. Pezzagna, J. Meijer, J. Du, C. A. Meriles, F. Reinhard, and J. Wrachtrup, *Science* **339**, 561 (2013).
- [6] I. Schwartz, J. Roskopf, S. Schmitt, B. Tratzmiller, Q. Chen, L. P. McGuinness, F. Jelezko, and M. B. Plenio, *Sci. Rep.* **9**, 6938 (2019).
- [7] N. Staudenmaier, A. Vijayakumar-Sreeja, S. Oviedo-Casado, G. Genov, D. Cohen, D. Dulong, T. Unden, N. Striegler, A. Marshall, J. Scheuer, C. Findler, J. Lang, I. Schwartz, P. Neumann, A. Retzker, and F. Jelezko, *npj Quantum Inf.* **8**, 120 (2022).
- [8] N. Staudenmaier, A. Vijayakumar-Sreeja, G. Genov, D. Cohen, C. Findler, J. Lang, A. Retzker, F. Jelezko, and S. Oviedo-Casado, *Phys. Rev. Lett.* **131**, 150801 (2023).
- [9] J. Du, F. Shi, X. Kong, F. Jelezko, and J. Wrachtrup, *Rev. Mod. Phys.* **96**, 025001 (2024).
- [10] A. O. Sushkov, I. Lovchinsky, N. Chisholm, R. L. Walsworth, H. Park, and M. D. Lukin, *Phys. Rev. Lett.* **113**, 197601 (2014).
- [11] Y. Wu, F. Jelezko, M. B. Plenio, and T. Weil, *Angew. Chem., Int. Ed. Engl.* **55**, 6586 (2016).
- [12] S. J. DeVience, L. M. Pham, I. Lovchinsky, A. O. Sushkov, N. Bar-Gill, C. Belthangady, F. Casola, M. Corbett, H. Zhang, M. Lukin *et al.*, *Nat. Nanotechnol.* **10**, 129 (2015).
- [13] A. Laraoui, F. Dolde, C. Burk, F. Reinhard, J. Wrachtrup, and C. A. Meriles, *Nat. Commun.* **4**, 1651 (2013).
- [14] T. Staudacher, N. Raatz, S. Pezzagna, J. Meijer, F. Reinhard, C. A. Meriles, and J. Wrachtrup, *Nat. Commun.* **6**, 8527 (2015).
- [15] X. Kong, A. Stark, J. Du, L. P. McGuinness, and F. Jelezko, *Phys. Rev. Appl.* **4**, 024004 (2015).
- [16] S. Zaiser, T. Rendler, I. Jakobi, T. Wolf, S. Y. Lee, S. Wagner, V. Bergholm, T. Schulte-Herbrüggen, P. Neumann, and J. Wrachtrup, *Nat. Commun.* **7**, 1 (2016).
- [17] M. Pfender, N. Aslam, H. Sumiya, S. Onoda, P. Neumann, J. Isoya, C. A. Meriles, and J. Wrachtrup, *Nat. Commun.* **8**, 1 (2017).
- [18] S. Schmitt, T. Gefen, F. M. Stürmer, T. Unden, G. Wolff, C. Müller, J. Scheuer, B. Naydenov, M. Markham, S. Pezzagna, J. Meijer, I. Schwarz, M. Plenio, A. Retzker, L. P. McGuinness, and F. Jelezko, *Science* **356**, 832 (2017).
- [19] J. M. Boss, K. S. Cujia, J. Zopes, and C. L. Degen, *Science* **356**, 837 (2017).
- [20] D. R. Glenn, D. B. Bucher, J. Lee, M. D. Lukin, H. Park, and R. L. Walsworth, *Nature (London)* **555**, 351 (2018).
- [21] N. Arunkumar, D. B. Bucher, M. J. Turner, P. TomHon, D. Glenn, S. Lehmkuhl, M. D. Lukin, H. Park, M. S. Rosen, T. Theis, and R. L. Walsworth, *PRX Quantum* **2**, 010305 (2021).
- [22] N. R. Neuling, R. D. Allert, and D. B. Bucher, *Curr. Opin. Biotechnol.* **83**, 102975 (2023).
- [23] T. Roskopf, J. Zopes, J. M. Boss, and C. L. Degen, *npj Quantum Inf.* **3**, 33 (2017).
- [24] J. F. Barry, J. M. Schloss, E. Bauch, M. J. Turner, C. A. Hart, L. M. Pham, and R. L. Walsworth, *Rev. Mod. Phys.* **92**, 015004 (2020).
- [25] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/1cj2-rxkm>, for details on the theory multipoint correlation spectroscopy and additional experimental results, which includes Refs. [26–59].
- [26] P. Neumann, J. Beck, M. Steiner, F. Rempp, H. Fedder, P. R. Hemmer, J. Wrachtrup, and F. Jelezko, *Science* **329**, 542 (2010).
- [27] T. Gullion, D. B. Baker, and M. S. Conradi, *J. Magn. Reson.* **89**, 479 (1990).
- [28] C. L. Degen, F. Reinhard, and P. Cappellaro, *Rev. Mod. Phys.* **89**, 035002 (2017).
- [29] D. Suter and G. A. Álvarez, *Rev. Mod. Phys.* **88**, 041001 (2016).
- [30] G. T. Genov, D. Schraft, N. V. Vitanov, and T. Halfmann, *Phys. Rev. Lett.* **118**, 133202 (2017).
- [31] G. T. Genov, Y. Ben-Shalom, F. Jelezko, A. Retzker, and N. Bar-Gill, *Phys. Rev. Res.* **2**, 033216 (2020).
- [32] D. Louzon, G. T. Genov, N. Staudenmaier, F. Frank, J. Lang, M. L. Markham, A. Retzker, and F. Jelezko, *Phys. Rev. Lett.* **134**, 120802 (2025).
- [33] J. M. Binder, A. Stark, N. Tomek, J. Scheuer, F. Frank, K. D. Jahnke, C. Müller, S. Schmitt, M. H. Metsch, T. Unden, T. Gehring, A. Huck, U. L. Andersen, L. J. Rogers, and F. Jelezko, *SoftwareX* **6**, 85 (2017).
- [34] D. Cohen, R. Nigmatullin, M. Eldar, and A. Retzker, *Adv. Quantum Technol.* **3**, 2000019 (2020).
- [35] K. S. Liu, X. Ma, R. Rizzato, A. L. Semrau, A. Henning, I. D. Sharp, R. A. Fischer, and D. B. Bucher, *Nano Lett.* **22**, 9876 (2022).
- [36] W. Zheng, S. Zhang, J. Jiang, Y. He, R. Stöhr, A. Denisenko, J. Wrachtrup, X. C. Zeng, K. Bian, E.-G. Wang *et al.*, *arXiv:2412.15001*.
- [37] R. Maier, C.-I. Ho, H. Sumiya, S. Onoda, J. Isoya, V. Vorobyov, and J. Wrachtrup, following Letter, *Phys. Rev. Lett.* **135**, 250802 (2025).
- [38] D. Suter and F. Jelezko, *Prog. Nucl. Magn. Reson. Spectrosc.* **98–99**, 50 (2017).
- [39] V. Jacques, P. Neumann, J. Beck, M. Markham, D. Twitchen, J. Meijer, F. Kaiser, G. Balasubramanian, F. Jelezko, and J. Wrachtrup, *Phys. Rev. Lett.* **102**, 057403 (2009).
- [40] A. M. Souza, G. A. Álvarez, and D. Suter, *Phil. Trans. R. Soc. A* **370**, 4748 (2012).
- [41] S. Oviedo-Casado, A. Rotem, R. Nigmatullin, J. Prior, and A. Retzker, *Sci. Rep.* **10**, 19691 (2020).
- [42] E. Hahn, *Phys. Rev.* **80**, 580 (1950).
- [43] Z.-Y. Wang, J. E. Lang, S. Schmitt, J. Lang, J. Casanova, L. McGuinness, T. S. Monteiro, F. Jelezko, and M. B. Plenio, *Phys. Rev. Lett.* **122**, 200403 (2019).
- [44] M. Garwood and Y. Ke, *J. Magn. Reson.* **94**, 511 (1991).
- [45] A. Tannús and M. Garwood, *NMR Biomed.* **10**, 423 (1997).
- [46] T. Joas, F. Ferlemand, R. Sailer, P. J. Vetter, J. Zhang, R. S. Said, T. Teraji, S. Onoda, T. Calarco, G. Genov, M. M. Müller, and F. Jelezko, *Phys. Rev. X* **15**, 021069 (2025).
- [47] D. Daems, A. Ruschhaupt, D. Sugny, and S. Guérin, *Phys. Rev. Lett.* **111**, 050404 (2013).

- [48] L. Van-Damme, D. Schraft, G. T. Genov, D. Sugny, T. Halfmann, and S. Guérin, *Phys. Rev. A* **96**, 022309 (2017).
- [49] M. H. Levitt, *Prog. Nucl. Magn. Reson. Spectrosc.* **18**, 61 (1986).
- [50] G. T. Genov, D. Schraft, T. Halfmann, and N. V. Vitanov, *Phys. Rev. Lett.* **113**, 043001 (2014).
- [51] P. Rembold, N. Oshnik, M. M. Müller, S. Montangero, T. Calarco, and E. Neu, *AVS Quantum Sci.* **2**, 024701 (2020).
- [52] H. M. Lim, G. T. Genov, R. Sailer, A. Fahrurrahman, M. A. Majidi, F. Jelezko, and R. S. Said, *Phys. Rev. Appl.* **24**, 054064 (2025).
- [53] J. Yoon, K. Zhong, G. Wang, B. Li, D. Lee, and P. Cappellaro, [arXiv:2504.12134](https://arxiv.org/abs/2504.12134).
- [54] Y.-X. Liu, A. Ajoy, and P. Cappellaro, *Phys. Rev. Lett.* **122**, 100501 (2019).
- [55] A. Jarmola, A. Berzins, J. Smits, K. Smits, J. Prikulis, F. Gahbauer, R. Ferber, D. Ertz, M. Auzinsh, and D. Budker, *Appl. Phys. Lett.* **107**, 242403 (2015).
- [56] P. J. Vetter, T. Reisser, M. G. Hirsch, T. Calarco, F. Motzoi, F. Jelezko, and M. M. Müller, *npj Quantum Inf.* **10**, 96 (2024).
- [57] P. J. Vetter, A. Marshall, G. T. Genov, T. F. Weiss, N. Striegler, E. F. Großmann, S. Oviedo-Casado, J. Cerrillo, J. Prior, P. Neumann *et al.*, *Phys. Rev. Appl.* **17**, 044028 (2022).
- [58] J. Fuhrmann, C. Findler, M. Olney-Fraser, L. Kazak, and F. Jelezko, *Adv. Funct. Mater.* 2501661 (2025), [10.1002/adfm.202501661](https://doi.org/10.1002/adfm.202501661).
- [59] B. Herzog, D. Cadeddu, F. Xue, P. Peddibhotla, and M. Poggio, *Appl. Phys. Lett.* **105**, 043112 (2014).
- [60] D. Wirtitsch, G. Wachter, S. Reisenbauer, M. Gulka, V. Ivády, F. Jelezko, A. Gali, M. Nesladek, and M. Trupke, *Phys. Rev. Res.* **5**, 013014 (2023).
- [61] D. A. Hopper, J. D. Lauigan, T.-Y. Huang, and L. C. Bassett, *Phys. Rev. Appl.* **13**, 024016 (2020).
- [62] L. M. Pham, S. J. DeVience, F. Casola, I. Lovchinsky, A. O. Sushkov, E. Bersin, J. Lee, E. Urbach, P. Cappellaro, H. Park *et al.*, *Phys. Rev. B* **93**, 045425 (2016).
- [63] T. Spohn, Plot data for manuscript: “Quantum memory enhanced multipoint correlation spectroscopy for statistically polarized NMR”, Zenodo, [10.5281/zenodo.17659892](https://zenodo.org/record/17659892) (2025).