

Multiplexed Scanning Microscopy with Dual-Qubit Spin Sensors

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Scanning probe microscopy with multiqubit sensors offers the potential to improve imaging speed and measure previously inaccessible quantities, such as two-point correlations. We develop a multiplexed quantum sensing approach with scanning probes containing two nitrogen-vacancy (NV) centers at the tip apex. A shared optical channel is used for simultaneous qubit initialization and readout, while phase- and frequency-dependent microwave spin manipulations are leveraged for demultiplexing the optical readout signal. Scanning dual-NV magnetometry is first demonstrated by simultaneously imaging multiple field projections of a ferrimagnetic racetrack device. Then, we record the two-point covariance of spatially correlated field fluctuations across a current-carrying wire. Our multiplex framework establishes a method to investigate a variety of spatiotemporal correlations, including phase transitions and electronic noise, with nanoscale resolution.

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The successes of quantum sensing with spin defects, particularly with nitrogen-vacancy (NV) centers in diamond, are supported by the high sensitivity, nanoscale resolution, and robust room-temperature functionality characteristic of these pointlike sensors [1,2]. When spin defects are integrated into scanning probe geometries [3,4], simultaneous topography and stray-field imaging becomes possible. Such experiments have led to substantial advances when investigating nanoscale magnetic textures [5], current distributions [6–8], superconductivity [9], and even ferroelectricity [10].

The majority of scanning probe studies rely on single-NV probes, owing to straightforward probe characterization and interpretation of measurement data. The relatively few investigations using multi-NV scanning probes [11–14] have yet to fully explore the potential benefits of operating multiple spin sensors at the same time. For example, spin ensembles confined in nanoscale volumes present the opportunity to improve measurement speed or sensitivity while achieving spatial resolution below the optical diffraction limit. Intriguing prospects also lie in imaging quantities inaccessible to a single sensor, such as spatial correlations

from field noise and fluctuations [15,16]. Two-point correlation measurements in other scanning probe microscopies [17–19] require multiple tips and complex experimental setups, however, with spin defects in diamond, a single probe can easily host two or more individual sensors. Moreover, methods are available to deterministically place defects within tens of nanometers [20].

In this Letter, we develop and realize scanning magnetometry experiments with diamond probes containing two active NV centers. Our concept uses a single optical channel for spin-state initialization and readout that is shared between NV centers, while phase-cycled microwave pulses allow for individual addressing of the two spins and demultiplexing of the optical output signal into individual qubit responses. Using a ferrimagnetic racetrack and a current-carrying wire as test devices, we demonstrate multiplexed imaging of magnetic field maps and covariance detection of spatially correlated field fluctuations with ~ 50 nm spatial resolution. The scanning probe geometry provides a unique test bed for investigating spatiotemporal correlations in general two-dimensional samples.

To introduce our sensing concept, we consider two qubits exposed to the same time-varying magnetic field $\vec{B}(\vec{r}, t)$, see Fig. 1. From the positions of each qubit, the vector field contains two contributions: a global contribution $\vec{B}_c(t)$ experienced by both qubits, and local contributions $\vec{B}_{uc,i}(t)$ unique to each qubit [Fig. 1(b)]. The field component $B_i(t)$ observed by the i th qubit is

$$B_i(t) = \vec{e}_i \cdot \left(\vec{B}_c(t) + \vec{B}_{uc,i}(t) \right), \quad (1)$$

where $\vec{e}_i$ [cf. Fig. 1(b)] is the projection axis of qubit i .

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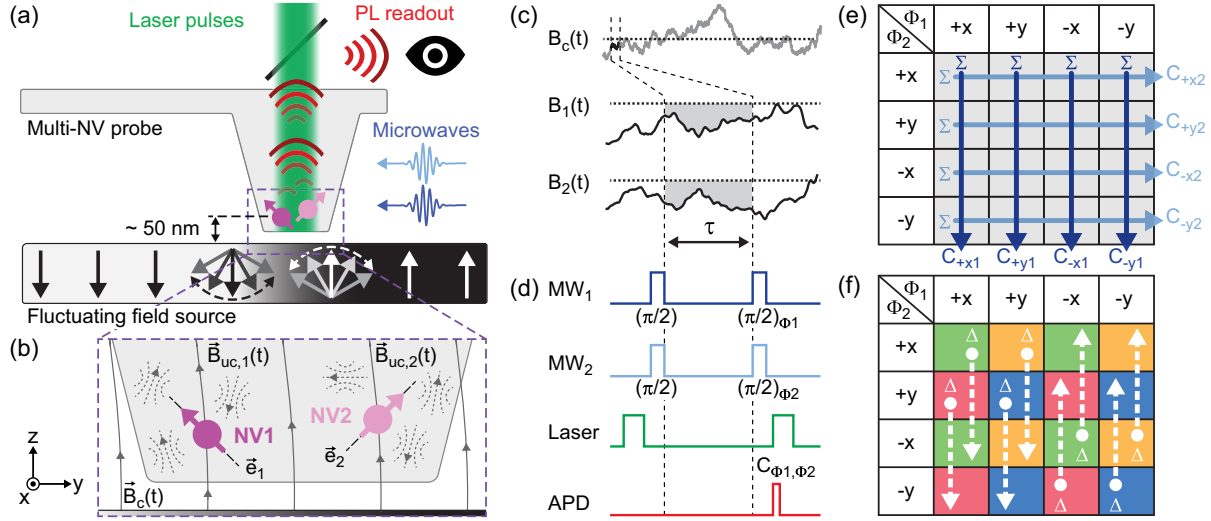


FIG. 1. Concept of dual-NV scanning microscopy. (a) Schematic of the scanning probe microscope. As the sample is moved under the probe, NV centers in the diamond tip sense their local magnetic (or electric) environment. A single optical channel is used for laser excitation and PL readout, while separate microwave frequencies control both spins. (b) Detail showing the correlated ($\vec{B}_c$, solid field lines) and uncorrelated ($\vec{B}_{uc,1}$, $\vec{B}_{uc,2}$, dashed) magnetic fields experienced by NV1 and NV2. The black-dashed lines indicate the axes $\vec{e}_i$. (c) Time trace of the global magnetic signal $\vec{B}_c(t)$ (arbitrary projection shown) and its projection onto every NV center (B_i). (d) Pulse sequence schematic. Laser pulses polarize NV centers and prompt PL emission. Fixed microwave frequencies (MW_i) simultaneously address a spin transition for each NV center. $\pi/2$ denotes the spin rotation angle and $\Phi_i \in \{+x, +y, -x, -y\}$ is the readout phase, representing the four qubit axes in the xy plane of the Bloch sphere [6,21]. An avalanche photodiode (APD) converts the PL emission into photon counts signals $C_{\Phi_1\Phi_2}$ that are used to recover the mean phases and count covariances. (e) Readout matrix for demultiplexing of mean phases $\bar{\phi}_1$ and $\bar{\phi}_2$. Each matrix element represents a photon count $C_{\Phi_1\Phi_2}$. Mean phases are obtained by partial summation (arrows) followed by application of Eq. (5). (f) Readout matrix for covariance sensing. Each matrix element represents the difference $V_{\Phi_1\Phi_2} - E_{\Phi_1\Phi_2}$, where V and E are the variance and expectation values, respectively. Covariances are obtained by subtracting the readout combination connected by dashed arrows. Four covariances can be computed, Cov_{xx} (green), Cov_{xy} (red), Cov_{yx} (yellow), Cov_{yy} (blue).

In our sensing experiment, the magnetic field is probed via the accumulated phase difference in a time-evolved superposition of states [22]. For a slowly varying magnetic field $B(t)$, the qubit-acquired phase is given by

$$\phi_i(t) = \gamma\tau B_i(t), \quad (2)$$

where τ is the phase accumulation time and $\gamma = 2\pi \times 28 \text{ GHz/T}$ the NV gyromagnetic ratio [1]. To better understand how the correlated and uncorrelated magnetic fields impact multisensor measurements, we express the readout phases of two NV centers as

$$\begin{aligned} \phi_1(t) &= \bar{\phi}_1 + \phi_c(t) + \phi_{uc,1}(t), \\ \phi_2(t) &= \bar{\phi}_2 + m\phi_c(t) + \phi_{uc,2}(t), \end{aligned} \quad (3)$$

where $m = (\vec{e}_2 \cdot \vec{B}_c) / (\vec{e}_1 \cdot \vec{B}_c)$. Derived in the Supplemental Material [23], Eq. (3) states that the total phases ϕ_i are composed of mean phases ($\bar{\phi}_i$), a correlated noise (ϕ_c and $m\phi_c$), and uncorrelated noises ($\phi_{uc,i}$). Together, m and ϕ_c characterize the strength of the fluctuations and the spatial correlation, which depends on the orientation and separations between the sensors.

Because the time-dependent terms average out across multiple measurements, only the mean phases $\bar{\phi}_i$ are usually measured in phased-based imaging experiments [6,8,10]. However, the noise term $\phi_c(t)$ generates a nonzero covariance between sensors accessible across many repeated measurements. This covariance signal contains important information about the spatial-temporal correlations of the magnetic field [15]. A scheme able to measure both the mean phases and signal correlations across multiple sensors would therefore be a valuable asset in the metrology of spatially correlated signals.

Our implementation for such a scheme is presented in Fig. 1(d). The pulse sequence uses two fixed frequencies—each tuned to the spin resonance of one NV center—applied at the same time [26]. Sixteen unique microwave phase readout combinations (denoted by Φ_1 and Φ_2) then permit separating both NV center contributions in the summed photoluminescence (PL) signal [27]. This readout scheme is capable of measuring both static fields [with the Ramsey protocol shown in Fig. 1(d)], coherent ac fields (with dynamical decoupling protocols [21,28,29]), and spatial correlations through covariance measurements (details below). Moreover, this scheme can be readily

generalized to more than two sensors [23]. The only requirement is that spin transitions are spectrally separated to minimize spurious excitation or dynamical frequency shifts [30]. When spin transitions for the sensors are degenerate, a subset of the sixteen-phase readout scheme can be used to sense the mean phase difference $|\bar{\phi}_1 - \bar{\phi}_2|$ [23].

After n measurement repetitions, the phase-dependent PL counts have the form

$$C_{\Phi_1\Phi_2} = nc_1 \left(1 - \frac{\epsilon_1}{2} \left[1 + e^{-\zeta_1(\tau)} \cos(\bar{\phi}_1 + \Phi_1) \right] \right) + nc_2 \left(1 - \frac{\epsilon_2}{2} \left[1 + e^{-\zeta_2(\tau)} \cos(\bar{\phi}_2 + \Phi_2) \right] \right), \quad (4)$$

where c_i , ϵ_i , $e^{-\zeta_i(\tau)}$ denote the average number of photons detected per readout for the $m_S = 0$ state, the optical contrast, and the spin dephasing for each NV center, respectively [21,23]. Typical values for our experiments are $c_i \sim 0.1$, $\epsilon_i \sim 0.2$, $\tau \sim 250$ ns, $\zeta_i(\tau) \sim 0.7$, and $n \sim 10^5$ – 10^8 .

The mean phases $\bar{\phi}_1$ and $\bar{\phi}_2$ can be extracted by combining partial sums of the PL counts, defined as $C_{\Phi_1} = \sum_{\Phi_2} C_{\Phi_1\Phi_2}$ and $C_{\Phi_2} = \sum_{\Phi_1} C_{\Phi_1\Phi_2}$ [blue arrows in Fig. 1(e)]. For the Ramsey protocol, the phase computation is

$$\phi_i = \arctan \left(\frac{C_{-y_i} - C_{+y_i}}{C_{-x_i} - C_{+x_i}} \right). \quad (5)$$

In Eqs. (4) and (5), each measurement repetition measures both NV centers and every phase combination is used to compute the accumulated mean phases. This establishes a multiplexed framework for simultaneously controlling and reading out multiple noninteracting qubits over a single optical channel.

To measure correlations, the Poissonian nature of the single-photon emission process during optical readout must be considered. Since the expected number of photons per readout E , and its variance V , are related to the phase accumulated, any covariance between NV centers [from ϕ_c and $m\phi_c$ in Eq. (3)], is found in the variance of the summed photon counts. The four readout phase combinations most sensitive to phase accumulation, $\Phi_1\Phi_2 = \pm y \pm y$, can be turned into a count covariance, $\text{Cov}_{yy} = \langle \sin \phi_1, \sin \phi_2 \rangle \sim \langle \phi_1, \phi_2 \rangle$, by combining the expectation values $E_{\pm y \pm y}$ and variances $V_{\pm y \pm y}$ taken across many readouts [23]. From the law of total covariance, one of the two equivalent expressions for Cov_{yy} is

$$\text{Cov}_{yy} = \frac{(V_{+y+y} - E_{+y+y}) - (V_{+y-y} - E_{+y-y})}{c_{r1}c_{r2}}. \quad (6)$$

Here, $c_{ri} = c_i \epsilon_i e^{-\zeta_i(\tau)}$ is the readout contrast of each NV center [23]. Three other count covariances (Cov_{xx} , Cov_{xy} ,

and Cov_{yx}) can be computed from the remaining sixteen-phase readouts through analogous expressions to Eq. (6), see Fig. 1(f) [23]. In general, all four count covariances are needed to isolate the covariance of the correlated phases ϕ_c and $m\phi_c$, since nonzero $\bar{\phi}_1$ and $\bar{\phi}_2$ mix the correlation among different count covariances [23]. However, when $\bar{\phi}_1 \approx \bar{\phi}_2 \approx 0$,

$$\text{Cov}_{yy} = \langle \sin(\phi_c), \sin(m\phi_c) \rangle \sim m\phi_c^2. \quad (7)$$

We experimentally demonstrate multiplexed and correlated sensing using the arrangement sketched in Fig. 1(a). Following typical scanning NV platforms [3,4], a sample is scanned underneath a diamond probe where laser pulses, microwave pulses, and single-photon counting are used for spin control and readout. Extending single-NV scanning microscopy to dual-NV (or multi-NV) microscopy therefore requires no change in instrumentation. Dual-NV probes (QZabre [31]) are fabricated by adjusting dopant concentrations during nitrogen implantation [32,33] and postselecting those containing two NV centers.

With two NV centers in the same nanophotonic structure (i.e., the diamond tip), laser irradiation interacts with both NV centers, coupling their photon emission and collection. When a laser is scanned across the probe, multiple bright regions are observed [Fig. 2(a)], reflecting the multimodal nature of these tips [34]. By contrast, a single-mode structure would only produce a single PL maximum regardless of the number of emitters. Because of the complex spatial structure of the laser mode volume, pillar shape, and random NV center location, attributing a bright spot in Fig. 2(a) to a certain NV center is not possible. Instead, each laser position excites one or both NV centers with varying efficiencies [23]. Positioning the laser focus to illuminate both NV centers [black cross in Fig. 2(a)] and sweeping the microwave excitation frequency reveals four PL dips in Fig. 2(b). These dips correspond to the two allowed spin transitions for each NV center [1,2,35]. Since the NV centers in this probe have different orientations ($\vec{e}_1 \nparallel \vec{e}_2$), a small magnetic bias field spectrally separates each transition.

In addition to having different orientations, NV centers also have different positions $\vec{r}_1$ and $\vec{r}_2$ as shown in Fig. 2(c). The spatial separation is between $0 - D$ in the xy plane, where $D \sim 300$ – 400 nm is the tip apex diameter [34], and ~ 5 – 20 nm in the z direction, owing to the ~ 5 keV ion implantation energy [33]. To properly interpret measurement data, the orientations and relative distance vector $\vec{r}_2 - \vec{r}_1 = (\Delta x, \Delta y, \Delta z)$ must be determined experimentally. To do so, we scan the tip across perpendicular edges of a magnetic calibration sample [36] and fit the ODMR profiles by a magnetic stray-field model [23]. This procedure gives estimates for the orientations of both NV centers, their lateral separation, and their respective z standoff distances. Figure 2(d) plots example line scan

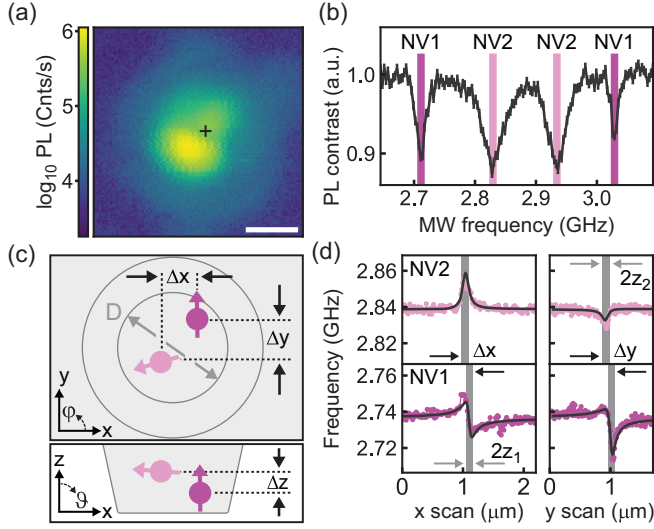


FIG. 2. Characterization of dual-NV probes. (a) Log-scale confocal image of the PL recorded by scanning the excitation laser across the diamond probe. Multiple bright spots indicate multiple NV centers. Scale bar, 1 μm . (b) Optically detected magnetic resonance (ODMR) spectra identifying four spin transitions, confirming the presence of two NV centers. The spectra are recorded while focusing the laser at the position indicated by the cross in (a). (c) Schematic of two NV centers showing their orientation (defined by ϑ and φ) and relative positions (defined by Δx , Δy , and Δz). (d) ODMR frequency shifts while scanning across orthogonal step edges of a magnetic stripe. Δx and Δy (black arrows) are determined by the fitted sample edges (gray lines). The gray line widths (indicated with gray arrows) are twice the fitted standoff distance ($z_1 = 47 \pm 1$ and $z_2 = 58 \pm 2$ nm for the plotted data), which determines Δz and the spatial resolution. Data recorded on tip No. 2.

data with fits for two of the four measured spin transitions. For the probe characterized in Fig. 2, the NV orientations are $\vec{e}_1 = (\vartheta_1, \varphi_1) = (41 \pm 2^\circ, 94 \pm 2^\circ)$ and $\vec{e}_2 = (\vartheta_2, \varphi_2) = (-84 \pm 2^\circ, -161 \pm 2^\circ)$, and their separation is $\Delta x = -52 \pm 7$ nm, $\Delta y = -96 \pm 6$ nm, and $\Delta z = 11 \pm 5$ nm in the laboratory frame. Characterization data for additional probes are given in [23].

We demonstrate multiplexed imaging on two representative magnetic samples. The first sample is a ferrimagnetic GdCo racetrack device [37] that is uniformly magnetized out of plane [23]. Images for the sixteen-phase readouts are shown in Fig. 3(a). Demultiplexing of the PL count images [Figs. 3(b), 3(c)] reproduces the individual phase maps $\bar{\phi}_1$ and $\bar{\phi}_2$, representing different projections of the stray field vector onto each NV center. In addition, the images are slightly shifted because the NV centers sit at different locations in the tip ($\sqrt{\Delta x^2 + \Delta y^2} \sim 180$ nm for this tip). The ability to simultaneously provide multiple vector field projections is one advantage of multiqubit probes [38]. Opposite to ensemble probes [14], however, our measurement scheme maintains the nanoscale spatial resolution offered by single spin probes. Additionally, experiments

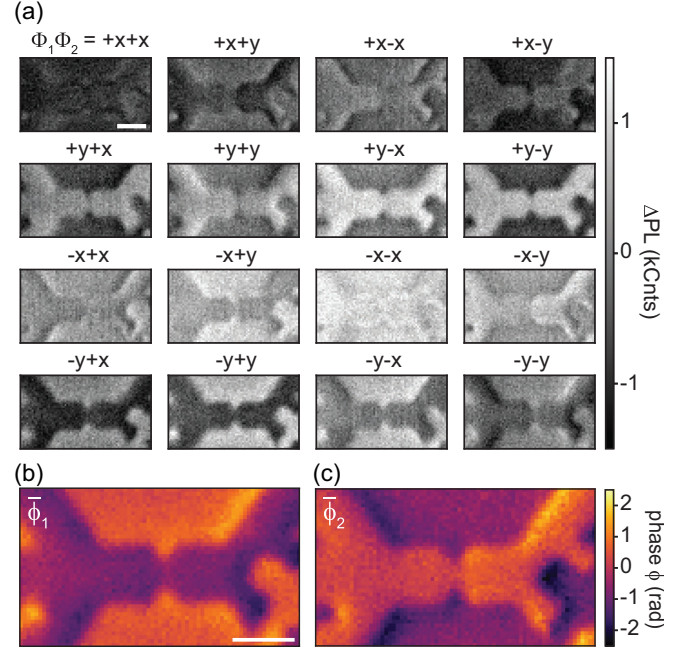


FIG. 3. Multiplexed imaging of a ferrimagnetic racetrack. (a) PL count data, $C_{\Phi_1 \Phi_2}$ with mean PL subtraction, for the sixteen readout combinations forming the matrix in Fig. 1(e). (b), (c) Reconstructed phase images $\bar{\phi}_1$ (NV1) and $\bar{\phi}_2$ (NV2). Scale bars, 2 μm . Data recorded on tip No. 4.

show that simultaneous operation of both NV centers improves measurement sensitivity by approximately $\sqrt{2}$, as expected from the $\propto \sqrt{N}$ scaling for N sensors [22,23].

For our second experiment we use a current-carrying wire [39] consisting of a ~ 700 nm wide Ti-Au stripe, see Fig. 4(a) [23]. Static and fluctuating magnetic fields are generated by injecting dc or asynchronous ac current waveforms. Figure 4(b) displays field profiles for dc injection recorded across the width of the wire. Similar to Figs. 3(b), 3(c), multiplexing is able to record separate profiles for NV1 and NV2, reflecting the differences in sensor orientation and position.

To demonstrate covariance imaging, we inject an ac current that runs asynchronous to the measurements sequence. By choosing a frequency f much slower than the duration of a single phase measurement ($f \sim 35$ kHz $\ll \tau^{-1} = 4$ MHz), individual phase readouts can be treated like random samples from an arcsine distribution. A correlated phase $\phi_c(t)$ is formed since the fluctuating current in the wire produces the fields observed by both NV centers.

Figures 4(c), 4(d) display the PL count data from three asynchronous ac scanning experiments. The phase contrasts c_{r1} and c_{r2} in Fig. 4(c) show dips of the root-mean-square (rms) magnetic noise observed by each NV center. Similar decoherence-based measurements are used in the detection of fluctuating spins [40,41]. The dips mimic the static field measurements in Fig. 4(b), since the rms noise

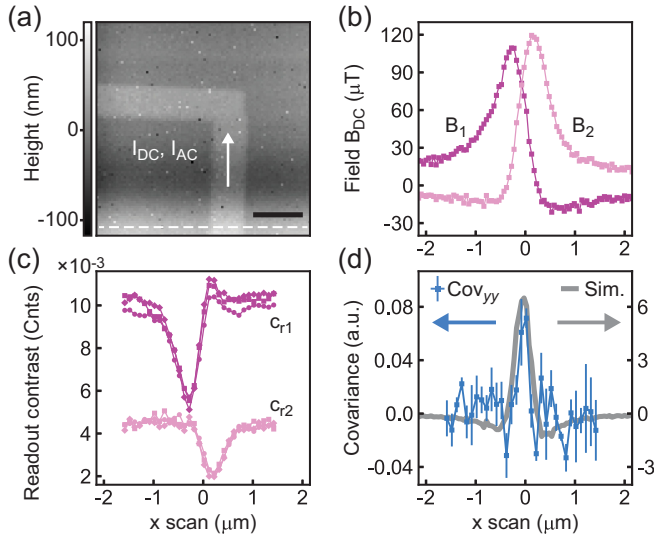


FIG. 4. Sensing spatial correlations with covariance detection. (a) Topography of the current-carrying wire. Scale bar, 1 μm . (b) Dual-NV magnetometry line scans taken across the wire [white-dashed line in (a)]. A dc current through the wire produces a static magnetic field. (c) Phase contrasts c_{r1} and c_{r2} for three covariance imaging measurements where an asynchronous ac current ($f = 35.211430$ kHz) is passed through the wire. Dips in the signal reflect magnetic noise seen by each NV center, leading to decoherence. Data point shapes (square, circle, and diamond) indicate different experimental runs. (d) Covariance signal computed from the averaged measurements of (c). Error bars are the standard deviation between the measurements. Gray curve is a Monte Carlo simulation of the expected covariance using the measurements in (b). Data recorded on tip No. 4.

of an oscillating signal is proportional to the amplitude of oscillation. Different background levels for c_{r1} and c_{r2} are due to changes in the laser focus position. The averaged count covariance, Cov_{yy} , is shown in Fig. 4(d). The singly peaked shape of the data, which corresponds to the region where both NV centers see strong fluctuations with the same polarity, agree well with the covariance profile estimated from the dc data (gray curve) [23]. With negligibly small static fields in the measurements, the entire correlated signal is found in the Cov_{yy} measurement.

The relatively low signal-to-noise ratio (SNR) in the covariance data [Fig. 4(d)] is due to a combination of low initialization and readout fidelities as well as the presence of uncorrelated noise causing additional decoherence, leading to $c_{ri} \sim 10^{-3} \ll 1$ [Fig. 4(c)]. Since the SNR depends quadratically on c_{ri} [15], improved laser initialization and readout protocols [42,43], photonic wave guiding [34,44] and charge-state or logic-based readouts [15,45,46] could strongly boost sensitivity. Substantial SNR gains could also be made by increasing τ through dynamical decoupling [47] since $\phi_c^2 \propto \tau^2$. Dynamical decoupling could further be used to decipher the frequency spectra of noise sources [48]. Alternatively, relative time shifts between microwave pulses could be introduced to

analyze temporal correlations at nanosecond timescales [49], offering perspectives for probing fast spatiotemporal correlations.

Looking forward, the ability to measure multiple qubits simultaneously, including the spatial correlation among qubits, creates the possibility to investigate a variety of correlated phenomena with multi-NV scanning probes. This includes spatial phase transitions in magnetic, superconducting, and ferroelectric materials [50–52], as well as correlated electronic noise in conductors [53,54], including hydrodynamic flow [55,56]. Additionally, while our experimental demonstration used two noninteracting qubits, our framework is scalable to more sensors in a single probe—up to four NV centers based on the crystallographic orientations and more by discriminating NV's by their individual ^{13}C nuclear spin environments [23,57]. Our approach can therefore serve as a starting point for imaging higher-order correlations [16] or for sensing with entangled states. The latter approach, where sensor separations $\lesssim 30$ nm would be needed [58], could enable sensing beyond the standard quantum limit [22,59].

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