

Bias-Field-Free Operation of Nitrogen-Vacancy Ensembles in Diamond for Accurate Vector Magnetometry

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Accurate measurement of vector magnetic fields is critical for applications including navigation, geoscience, and space exploration. Nitrogen-vacancy (NV) center spin ensembles offer a promising solution for high-sensitivity vector magnetometry, as their different orientations in the diamond lattice measure different components of the magnetic field. However, the bias magnetic field typically used to separate signals from each NV orientation introduces inaccuracy from drifts in permanent magnets or coils. Here, we present a novel bias-field-free approach that labels the NV orientations via the direction of the microwave (MW) field in a variable-pulse-duration Ramsey sequence used to manipulate the spin ensemble. Numerical simulations demonstrate the possibility to isolate each orientation's signal with subnanotesla accuracy in most terrestrial fields, even without precise MW field calibration, at only a moderate cost to sensitivity. We also provide proof-of-principle experimental validation, observing relevant features that evolve as expected with the applied magnetic field. Looking forward, by removing a key source of drift, the proposed protocol lays the groundwork for future deployment of NV magnetometers in high-accuracy or long-duration missions.

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I. INTRODUCTION

Magnetic sensor development often focuses on precision, the ability to sense as small a change as possible. Nevertheless, in some contexts, accuracy or long-term stability can prove more important. Accurate maps of Earth's magnetic field are critical to navigation [1–4], geoscience [5–8], and resource exploration [9–11]; sensors that retain calibration over months or years are essential to exploratory space missions [12]. While scalar atomic magnetometers can exhibit absolute accuracy down to ~ 50 pT [13,14], vector mode operation remains a challenge, with field-deployed vector-mode atomic magnetometers reaching only approximately nanotesla accuracy [15,16]. Even hybrid solutions using a scalar atomic magnetometer for *in situ* calibration of a vector fluxgate sensor attain only slightly better accuracy, with the additional cost of

complex calibration procedures and reduced performance in high field gradients [17,18].

Nitrogen-vacancy (NV) centers in diamond offer an appealing alternative for accurate vector magnetometry because they combine the long spin coherence time and Hamiltonian-defined response of atomic magnetometers with intrinsic vector measurement capabilities arising from their site symmetry [19,20]. Each of the four possible NV center orientations in the diamond lattice [see Fig. 1(a)] is most sensitive to fields along its symmetry axis, permitting vector field measurement. To reconstruct the magnetic field, one must know which signals come from which NV orientation. The most common solution applies a bias field of a few milliteslas using permanent magnets or coils to separate the spin transition frequencies of different orientations, allowing orientation-labeled measurement of small shifts due to the external magnetic field [21,22]. This approach has enabled NV magnetometers with sufficient accuracy for selection as a finalist in the MagQuest Challenge [23,24], signaling the potential for NV magnetometers to outclass existing magnetometers for Earth field mapping. However, at the nanotesla accuracy level, the bias field becomes problematic because it is indistinguishable from the external field, such that drifts in bias due to thermal [25], mechanical, or hysteresis [26] effects induce systematic errors. Removing large bias fields could

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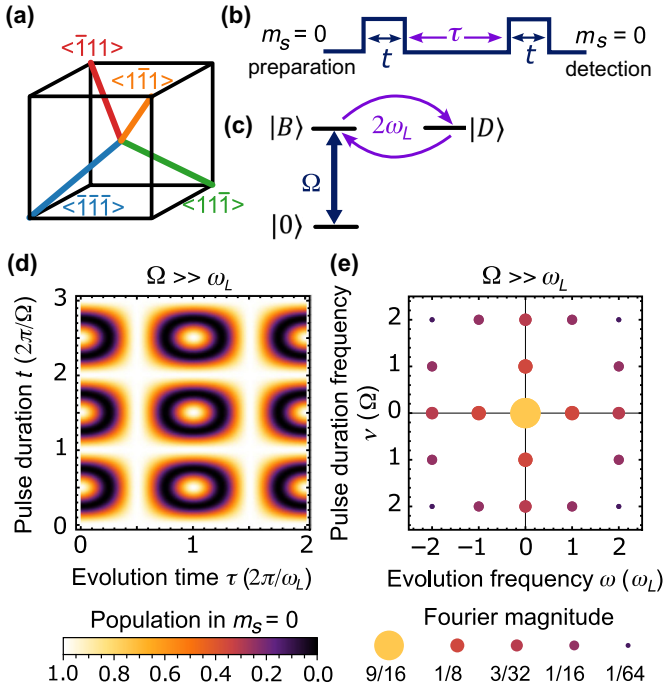


FIG. 1. (a) The four possible directions of the NV symmetry axis (N-to-V axis) in the conventional diamond crystal unit cell. (b) Variable-pulse-duration Ramsey sequence with two microwave pulses of duration t separated by free evolution time τ . (c) Dynamics in the bright-dark basis. (d) Population in $|0\rangle$ ($P_0(t, \tau)$) after application of the pulse sequence in (b) in the $\Omega/\omega_L \rightarrow \infty$ limit, with constant microwave phase, no detuning, no dephasing, and an axial magnetic field. The pulse duration t is given in units of the Rabi period and the evolution time τ is given in units of the Larmor period. (e) Magnitude of the two-dimensional Fourier transform of the population in (d), with discrete frequencies illustrated with dots of area proportional to their magnitude. The quantitative values given in the legend are $|a_{nm}|$ in the Fourier expansion $P_0(t, \tau) = \sum_n \sum_m a_{nm} e^{i\nu n t} e^{i\omega m \tau}$. The frequency associated with the pulse duration (free evolution time) is given in units of the Rabi frequency Ω (Larmor frequency ω_L).

also aid materials research applications, as millitesla fields are incompatible with some target samples [27]. Bias-field-free operation is thus desirable to enable high-accuracy NV vector magnetometry and expand its range of applications.

Indeed, there has been a recent surge of interest in bias-field-free schemes. Several groups have explored the use of optical anisotropy, labeling orientations via distinct spatial emission patterns [28–30] or excitation and emission polarization [31–34]. When applied to NV ensembles, however, these schemes incompletely isolate the contributions from different orientations, such that any drifts in the optical calibration will degrade accuracy, particularly when two orientations experience similar magnetic field projections such that their spin transitions nearly coincide and relevant features overlap. Microwave (MW) polarization has also been suggested to discern vector

fields from a single orientation [35] or label orientations, in the latter case either on its own [36] or combined with optical polarization [32]. These proposed implementations using MW polarization exhibit accuracy issues similar to those of optical polarization schemes. While circular MW polarization in combination with optimal control could in principle provide better isolation of each orientation’s signal [37], robust implementation over a terrestrial-magnetic-field range remains to be demonstrated. Pulsed biasing [38] could improve accuracy if a sufficiently precise control system can be realized, but it would not function for samples requiring a low field. Thus, it remains an open challenge to identify a robust, crosstalk-free, drift-free mechanism for labeling NV orientations in the absence of a bias field.

Here, we examine a novel method to label NV orientations via the MW field. By appropriate adjustment of the MW field angle relative to the diamond crystalline axes, each NV orientation experiences a distinct Rabi frequency. While Rabi labeling has been proposed before [39], we identify a technique that permits effective use of the Rabi-frequency label without relying on knowledge of the exact MW direction or a calibrated model of the NV spin resonance line shapes. In particular, we use a Ramsey-like protocol with MW pulses of variable duration [see Fig. 1(b)], separating the labeling dimension (pulse duration) from the sensing dimension (interpulse delay) to enable true isolation of signals originating from each orientation. Crucially, the variable-pulse-duration Ramsey (VPDR) protocol allows us to identify drifts in the MW field separately from changes in the target dc field, such that precise calibration of the MW magnitude and direction is not required. In addition, the VPDR sequence probes double-quantum (DQ) spin transitions, which increases stability in the presence of temperature fluctuations and eliminates dephasing from axial strain inhomogeneities [20]. The scheme requires only a single MW frequency, easing experimental implementation; while it cannot operate at true zero field, for reasonable parameters it has a range exceeding requirements for sensing in terrestrial fields.

The paper is structured as follows. In Sec. II we analytically model the response of a single NV center to the VPDR pulse sequence and analyze it in the frequency domain. This model provides insight into different regimes of operation and allows estimation of the sensitivity cost relative to more standard measurements. We then test the VPDR protocol on numerical simulations of an ensemble of NV spins in Sec. III, with open-source code for the simulator provided [40]. We demonstrate that it is possible to suppress crosstalk between signals from different orientations to reach subnanotesla accuracy across an Earth-magnetic-field range, apart from discrete dead zones that we identify and discuss. Moreover, we show that similar accuracy is retained even for factor-of-2 changes in

MW magnitude and $\pm 4^\circ$ MW direction fluctuations, such that accuracy is not constrained by MW calibration precision. Finally, in Sec. IV we show a proof-of-principle experimental demonstration, comparing observed features with those expected from the analytic theory, illustrating the feasibility of Rabi-labeled, bias-field-free vector field measurements.

II. THEORETICAL PRINCIPLES

A. Single NV center response

The VPDR pulse sequence [see Fig. 1(b)] comprises two MW pulses of duration t separated by a free evolution time τ , where the second MW pulse has a phase shift ϕ of 0° or 180° with respect to the first. We begin by analyzing the effect of the VPDR sequence on a single NV center modeled as a spin $S = 1$ system with a zero-field splitting $\Delta/(2\pi)$ that we set to 2.87 GHz [41]. We assume that the NV spin is initially polarized into the $m_s = 0$ Zeeman projection ($|0\rangle$), and that a small axial field B_z (to be measured) splits the $m_s = \pm 1$ states ($|\pm 1\rangle$) by energy $2\hbar\omega_L = 2g\mu_B B_z$. A MW field $\mathbf{B}_{\text{MW}} \cos(\nu_{\text{MW}}t + \phi)$ is applied to the spin. By choosing the $\hat{z}$ coordinate axis along the NV symmetry axis and the $\hat{x}$ coordinate axis along the projection of the MW field in the plane perpendicular to $\hat{z}$ ($\mathbf{B}_{\text{MW},\perp}$), we can write the NV spin Hamiltonian in the rotating frame and rotating wave approximation as

$$H = -\hbar\delta\hat{S}_z^2 + \hbar\omega_L\hat{S}_z + \frac{\hbar\Omega}{2} \left(\cos\phi\hat{S}_x + \sin\phi(\hat{S}_y\hat{S}_z + \hat{S}_z\hat{S}_y) \right), \quad (1)$$

where $\Omega = g\mu_B B_{\text{MW},\perp}/\hbar$ is the Rabi frequency, $\delta = \nu_{\text{MW}} - \Delta$ is the MW detuning from the zero-field splitting (unless otherwise specified, we assume $\delta = 0$), and $\hat{S}_x$, $\hat{S}_y$, and $\hat{S}_z$ are the spin-1 matrices. Note that the MW phase ϕ is not analogous to the MW direction for a spin-1 system.

1. The high-Rabi-frequency limit

By propagating a $|0\rangle$ -initialized spin through the three intervals of the pulse sequence, we can obtain an expression for the final $m_s = 0$ population $P_0(t, \tau)$ (see Appendix A). In the hard-pulse limit $\Omega/\omega_L \rightarrow \infty$ and $\Omega/\delta \rightarrow \infty$,

$$\lim_{\substack{\Omega/\omega_L \rightarrow \infty \\ \Omega/\delta \rightarrow \infty}} P_0(t, \tau) \approx \cos^4 \frac{\Omega t}{2} + \underbrace{\frac{\cos(\tau\delta + \phi)}{2} \sin^2 \Omega t \cos \omega_L \tau}_{\text{SQ}} + \underbrace{\sin^4 \frac{\Omega t}{2} \cos^2 \omega_L \tau}_{\text{DQ}}. \quad (2)$$

An illustration of this signal for $\delta = 0, \phi = 0$ is shown in Fig. 1(d). From examination of Eq. (2), two of its terms exhibit oscillations with τ that can be used to deduce the Larmor precession frequency (and thus the magnetic field). The second term oscillates at ω_L [a single-quantum (SQ) Ramsey signal], while the third term oscillates at $2\omega_L$ (a DQ Ramsey signal).

Only single-quantum terms are sensitive to the detuning or phase of the microwaves. In particular, changing the phase of the second pulse by 180° reverses the sign of the SQ term, similarly to techniques used for double-quantum Ramsey in a bias field [42]. Thus, by two applications of the pulse sequence, once with $\phi = 0^\circ$ and once with $\phi = 180^\circ$, and addition of the resulting signals, the SQ contribution can be canceled. Isolating the DQ signal is beneficial because of its insensitivity to detuning (and thus to variations in temperature and axial strain); moreover, DQ transitions do not suffer from dephasing due to axial strain inhomogeneities, offering an opportunity for improved magnetic sensitivity [43–46].

Because $P_0(t, \tau)$ is a sum of oscillations, it is natural to examine its Fourier transform $\mathcal{P}_0(\nu, \omega) = \int dt \int d\tau e^{-i\nu t} e^{-i\omega \tau} P_0(t, \tau)$. In the absence of dephasing, $\mathcal{P}_0(\nu, \omega)$ is a sum of Dirac delta functions; Fig. 1(e) shows $\mathcal{P}_0(\nu, \omega)$ in the $\Omega/\omega_L \rightarrow \infty$ limit, with the magnitude of the area under each two-dimensional (2D) delta function represented by the area of the circle centered at its frequencies (ν, ω) . This Fourier domain representation enables a simple conceptual interpretation of data: the vertical location of a Fourier peak shows the Rabi frequency (which will be used to label the orientation), while its horizontal location indicates the free precession frequency associated with that Rabi frequency, thus revealing information about the projection of the magnetic field on that Rabi-labeled NV axis.

For quantitative field estimation, we will focus on the DQ term at $\nu = \Omega, \omega = 2\omega_L$. The origin of this term can be understood in the bright state–dark state picture [see Fig. 1(c)]. The pulsed MW drive couples $|0\rangle$ to the so-called bright state $|B\rangle = \frac{1}{\sqrt{2}}(|+1\rangle + |-1\rangle)$, producing oscillations between $|0\rangle$ and $|B\rangle$ at the Rabi frequency Ω . During free evolution, the axial magnetic field induces oscillations between $|B\rangle$ and the orthogonal dark state $|D\rangle = \frac{1}{\sqrt{2}}(|+1\rangle - |-1\rangle)$ at twice the Larmor precession frequency $2\omega_L$. Whenever the pulse duration t is an odd integer multiple of π/Ω , each pulse will transfer all the population into or out of the $\{|B\rangle, |D\rangle\}$ subspace, leading to a final $|0\rangle$ population that oscillates strongly with τ ; whenever t is an even multiple of π/Ω , the $\{|B\rangle, |D\rangle\}$ subspace will not be populated during the free evolution period and the Larmor precession signal vs τ will be suppressed. We thus expect the final $m_s = 0$ population to contain an oscillation with τ at frequency

$2\omega_L$ modulated by an oscillation with t at the Rabi frequency Ω .

It is straightforward to extract this DQ term of interest from a dataset acquired at a range of pulse durations t and one or more free evolution times τ . By taking the inner product of $P_0(t, \tau)$ with $\cos \Omega t$ along the pulse-duration dimension, one obtains the double-quantum Ramsey signal

$$\lim_{\substack{\Omega/\omega_L \rightarrow \infty \\ \Omega/\delta \rightarrow \infty \\ T \rightarrow \infty}} \frac{1}{T} \int_0^T 2 \cos \Omega t P_0(t, \tau) dt = \frac{1}{4} (1 - \cos 2\omega_L \tau), \quad (3)$$

from which the magnetic field along the NV axis can be inferred.

The theoretical sensitivity of the VPDR protocol is negatively impacted by taking measurements at pulse durations t that are not optimal. To quantify the sensitivity cost, we compare the VPDR protocol (performed for a given free evolution time τ) to a more standard DQ Ramsey experiment realized by applying two pulses of duration $t = \pi/\Omega$ separated by the same free evolution time τ , which would produce a $m_s = 0$ population $\cos^2 \omega_L \tau$. For VPDR data in the hard-pulse limit, analyzed via a cosine inner product on the pulse-duration dimension, sampling at many pulse durations worsens the sensitivity by only a factor of $2\sqrt{2}$ relative to the DQ Ramsey experiment. While this result holds even in the presence of dephasing during free evolution, it is valid only in the high-Rabi-frequency limit. For moderate Rabi frequencies, dephasing during MW pulses can further degrade VPDR sensitivity (see Appendix B for details).

2. Finite Ω/ω_L considerations

The simple expression in Eq. (2) is valid only in the $\Omega/\omega_L \rightarrow \infty, \Omega/\delta \rightarrow \infty$ limit. While we can impose $\delta = 0$, it is important to understand the effect of finite Ω/ω_L . The Larmor precession of the spin during the pulse modifies the response of the system in two ways: First, the $|0\rangle \rightarrow |B\rangle$ population oscillations occur at the effective Rabi frequency $\Omega_{\text{eff}} = \sqrt{\Omega^2 + 4\omega_L^2}$; secondly, new oscillations with t emerge at $\nu = \pm\Omega_{\text{eff}}/2$ and $\nu = \pm 3\Omega_{\text{eff}}/2$. The full analytic expressions for $\delta = 0$ are given in Appendix A, and the resulting signals can be understood conceptually in the Fourier domain as illustrated in Fig. 2(a). Note that ν is given in units of Ω_{eff} (not Ω), while ω is given in units of ω_L . Figure 2(a) clearly shows the emergence of new Fourier peaks, as well as changes to the magnitudes of the original peaks seen in Fig. 1(e).

Perhaps surprisingly, the new Fourier components persist to fairly large values of Ω/ω_L . Figure 2(b) shows the Fourier amplitudes of the DQ terms in the first quadrant, as indicated by arrows in Fig. 2(a), as a function of

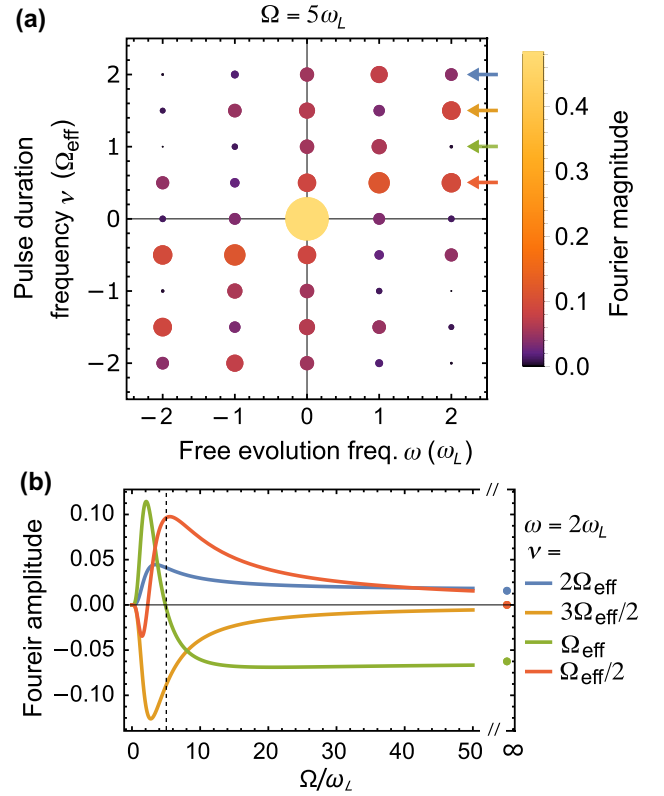


FIG. 2. (a) $\mathcal{P}_0(\nu, \omega)$ for $\Omega = 5\omega_L$, no detuning or dephasing, and constant microwave phase. The area of the dots is proportional to the magnitude of the discrete Fourier components as in Fig. 1(e). The pulse duration frequency ν is given in units of the effective Rabi frequency $\Omega_{\text{eff}} = \sqrt{\Omega^2 + 4\omega_L^2}$, while the free evolution frequency is given in units of the Larmor frequency ω_L . (b) Amplitude of the positive-quadrant double-quantum Fourier components, corresponding to the four components indicated with arrows in (a), as a function of Ω/ω_L . The dotted line indicates the value of Ω/ω_L for which the Fourier components in (a) are evaluated.

Ω/ω_L . In particular, the $\nu = \Omega_{\text{eff}}/2$ term remains appreciable even at $\Omega/\omega_L = 50$. In contrast, the term of interest at $\nu = \Omega_{\text{eff}}$ (green line) approaches its saturating value for $\Omega/\omega_L \gtrsim 10$. This behavior implies that analysis will be simpler when Ω exceeds ω_L by an order of magnitude or more.

Despite the additional signal complexity at finite Ω/ω_L , it remains possible to cancel the SQ peaks at $\omega = \omega_L$ by adding 0° - and 180° -phase-shifted signals (see Appendix A). Since the DQ signals are unaffected by the MW phase, this SQ cancellation procedure does not impact sensitivity, but implies a refresh rate reduction by a factor of 2.

3. Dephasing

Spin dephasing does not substantially change the interpretation of VPDR signals. In an analytic approximation

valid for $\Omega \gg \omega_L$, we ignore spin dephasing during the short pulses and focus on dephasing during the longer free evolution period. Modeling of the environmental noise as Lorentzian-distributed inhomogeneous variations in the local magnetic field [47] produces an exponential decay of all SQ signals as $e^{-\tau/T_2^*}$ and all DQ signals as $e^{-2\tau/T_2^*}$ (see Appendix A for details). In the high-Rabi-frequency approximation of Eq. (2), an inner product of the dephased signal with $\cos(\Omega t)$ yields $1/T \int_0^T 2 \cos \Omega t P_0(t, \tau) dt \approx \frac{1}{4} \left(1 - e^{-2\tau/T_2^*} \cos 2\omega_L \tau \right)$, which is half of the signal from a standard double-quantum Ramsey experiment, with the same decay term. As long as the dephasing during the pulses is negligible, a similar post-inner-product result can be obtained even at finite Ω/ω_L ; however, SQ cancellation will be necessary, and the magnitude of the DQ signal will vary with Ω/ω_L . Dephasing during the MW pulses does not lend itself to simple analytic treatment; as shown numerically in Appendix B, it further reduces the amplitude of the post-inner-product signal, worsening sensitivity but not impacting the functional dependence on τ (nor the resulting accuracy).

4. Hyperfine structure

Finally, NV centers have hyperfine structure associated with the host nitrogen nuclear spin. We consider the dominant $I = 1$ isotope ^{14}N , as the $I = \frac{1}{2}$ ^{15}N isotope would introduce additional modulation due to its lack of quadrupole splitting [48]. In the secular approximation, the ^{14}N nuclear spin adds an axial magnetic field proportional to its projection on the NV symmetry axis. When the nuclear spin is in Zeeman sublevel m_I , the effective Larmor frequency is $\omega_L = \omega_L^{\text{ext}} + m_I A$, where ω_L^{ext} is the Larmor precession frequency associated with an external axial magnetic field, and $A/(2\pi) = 2.16$ MHz [49]. Since the nuclear spin sublevels are approximately equally populated at room temperature and low magnetic field, we can account for hyperfine structure by averaging $P_0(t, \tau)$ over the three possible values of m_I . Qualitatively, hyperfine structure leads to a tripling of the Fourier-domain peaks in $\mathcal{P}(\nu, \omega)$ along the horizontal ω axis, corresponding to $\omega = m(\omega_L^{\text{ext}} + m_I A)$ with $m = \{-2, -1, 0, 1, 2\}$ and $m_I = \{-1, 0, 1\}$. With this additional complication, it becomes especially beneficial to cancel the SQ contribution by adding a 180° -phase-shifted signal, such that only $m = \{-2, 0, 2\}$ Fourier components are present in the signal. For the remaining theoretical and numerical analysis, we will work with such SQ-canceled signals.

Hyperfine structure also induces variation of the effective Rabi frequency with nuclear spin projection, $\Omega_{\text{eff}}(m_I) = \sqrt{\Omega^2 + 4(\omega_L^{\text{ext}} + m_I A)^2}$. Extraction of all three hyperfine signals from an inner product with a single cosine requires $\Omega \gg A$, further motivating the use of high Rabi frequency in an experimental realization.

B. Ensemble signals

When all four orientations of NV centers are present, each species i will have its own Rabi frequency $\Omega_i = (g\mu_B/\hbar)|\mathbf{B}_{\text{MW}} \times \hat{z}_i|$ set by the perpendicular projection of the MW magnetic field $\mathbf{B}_{\text{MW}}$ on the i th symmetry axis $\hat{z}_i$. Figure 3(a) shows an example of the Fourier transform of a numerically calculated ensemble VPDR signal (see Sec. III) with SQ cancellation for a magnetic field $|B_{\text{dc}}| \approx 0.35$ mT slightly canted from the $\langle 111 \rangle$ direction. As expected, the signals coming from the $\langle 111 \rangle$ subensemble occur at significantly higher free evolution frequency than signals from the other three orientations. Within the first quadrant, each orientation i exhibits 12 DQ Fourier components occurring at $\nu = n\Omega_{\text{eff},i}$ for $n \in \{\frac{1}{2}, 1, \frac{3}{2}, 2\}$ and $\omega = 2|\omega_{L,i}^{\text{ext}} + m_I A|$ for $m_I \in \{-1, 0, 1\}$. The bare Rabi frequencies Ω_i (as well as their harmonics) are illustrated with arrow-tipped horizontal lines, identifying each orientation.

A few features of Fig. 3(a) are worth noting. Owing to the large B_{dc} value, $\Omega_{\text{eff},i}$ differs from Ω_i , especially for the $\langle 111 \rangle$ orientation. The relatively small value of $\Omega_i/(\omega_{L,i}^{\text{ext}} + m_I A)$ is also responsible for the weak magnitude of the $\langle 111 \rangle$ peaks at $\nu = \Omega_{\text{eff},\langle 111 \rangle}$. Both these effects would be strongly suppressed for smaller B_{dc} within the range of Earth's field. More importantly, Fig. 3(a) demonstrates an advantage of the VPDR protocol over previous polarization-based approaches to bias-field-free orientation labeling [28,31,32]. For a field direction near $\langle 111 \rangle$, the three off-axis orientations have nearly commensurate spin transition frequencies, such that their optically detected magnetic resonance (ODMR) features would overlap. By separating the orientations' signals along the pulse duration dimension, the VPDR protocol ensures that their transition frequencies can be independently measured.

To fully isolate each orientation's signal, the Rabi frequencies Ω_i should be well separated, such that, for example, taking an inner product of the signal with $\cos \Omega_i t$ picks out the DQ signal from orientation i with minimal crosstalk from other orientations. Qualitatively, the best results will be obtained when the orientations' Rabi frequencies and their harmonics [arrows in Fig. 3(a)] do not come too close to each other, which can be controlled with the MW direction. We optimize the MW direction by calculating the smallest separation $|\Omega_i - n\Omega_j|$ for $i \neq j$ with $n \in \{\frac{1}{2}, 1, \frac{3}{2}, 2\}$ while varying the polar and azimuthal angles of $\mathbf{B}_{\text{MW}}$ relative to the conventional diamond unit cell. Figure 3(b) shows a plot of this smallest separation expressed as a percentage of the maximum Rabi frequency $\Omega_{\text{max}} = (g\mu_B/\hbar)|B_{\text{MW}}|$. Minimum separations just over 8% of Ω_{max} are possible, but they are achieved by making one of the Ω_i much smaller than the others, which could be problematic for experiments with limited Ω_{max} . The local maximum near $\theta_{\text{MW}} = 13.74^\circ$, $\phi_{\text{MW}} = 30.05^\circ$ with 6.8%

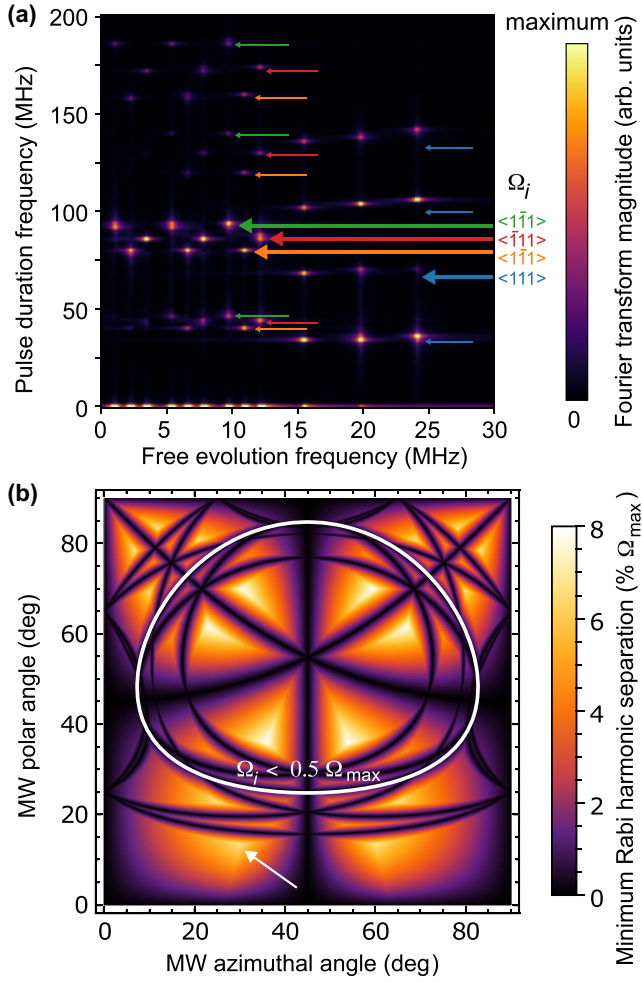


FIG. 3. (a) Discrete Fourier transform (positive-frequency quadrant only) of a simulated Variable-pulse-duration Ramsey signal with single-quantum cancellation for a NV ensemble. $\Omega_{\max}/2\pi = 100$ MHz, $T_2^* = 2$ μ s, $B = (185, 204, 223)$ μ T in the conventional diamond crystal coordinates, and $\mathbf{B}_{\text{MW}} \parallel (0.2054, 0.1188, 0.9714)$ corresponding to the locally optimal orientation indicated in (b). Thick arrow-tipped lines indicate each orientation's bare Rabi frequency Ω_i , while thin arrow-tipped lines indicate $\Omega_i/2$, $3\Omega_i/2$, and $2\Omega_i$. (b) Optimization of the microwave magnetic field direction. The minimum separation between Ω_i and $n\Omega_j$ for NV orientations $i \neq j$ and $n \in \{\frac{1}{2}, 1, \frac{3}{2}, 2\}$ is given as a function of the direction of the microwave magnetic field $\mathbf{B}_{\text{MW}}$ relative to the conventional diamond unit cell. For directions inside the white contour, one of the Ω_i is less than $\Omega_{\max}/2$; the white arrow indicates an optimal orientation outside the contour.

minimum separation (white arrow) may prove a better candidate because all four Ω_i exceed $0.65\Omega_{\max}$. For Fig. 3(a) and the numerical analysis in Sec. III, we use this MW orientation.

In an ensemble, off-axis dc magnetic fields are generally present for each NV orientation. The off-axis components of the field will partially hybridize the eigenstates of

the free-evolution Hamiltonian and shift their eigenvalues. In particular, the evolution with τ will now occur at the frequency difference $\Delta\omega_i$ between the $m_s = \pm 1$ -like eigenstates of the i th orientation. Nevertheless, it is meaningful to extract the $\Delta\omega_i$ as they can be used to reconstruct the magnetic field via least-squares fitting [22,50], linearized Hamiltonian inversion [51], or machine learning techniques [52]. Indeed, vector magnetometers based on continuous-wave ODMR also measure eigenvalue differences, so when evaluating the performance of the VPDR protocol, we will focus on accurately extracting the $\Delta\omega_i$ values. While the effects of off-axis fields are expected to be negligibly small in terrestrial fields [suppressed by $\sim (g\mu_B B_{\perp}/\hbar\Delta)^2$], we include them in the numerical simulations discussed below.

III. NUMERICAL SIMULATIONS

Our motivation for the VPDR protocol is the elimination of bias magnets for reduced drift and increased accuracy. It is thus essential to evaluate (1) the feasibility of accurately extracting $\Delta\omega_i$ from VPDR data without crosstalk from other orientations and (2) the robustness of this inversion regarding drifts in MW field amplitude or orientation. To these ends, we numerically simulate the response of an ensemble of NV centers. After projecting the external dc and MW magnetic fields onto each of the four possible NV coordinate systems, we solve the master equation for each spin species (see Appendix C for calculation details), including off-axis magnetic fields, Markovian dephasing throughout the pulse sequence, and hyperfine interactions in the secular approximation; the code is publicly available [40]. The simulation returns a SQ-canceled signal

$$S_{\text{VPDR}}(t, \tau) = P_0^{\text{total}}(t, \tau, \phi = 0) + P_0^{\text{total}}(t, \tau, \phi = \pi),$$

where $P_0^{\text{total}}(t, \tau, \phi)$ is the probability for an $m_s = 0$ -initialized spin in the ensemble to end up in $|0\rangle$ after a VPDR pulse sequence with relative MW pulse phase ϕ , averaged over NV orientation and nuclear spin projection. In particular, $S_{\text{VPDR}}(t, \tau)$ should be proportional to the sum of signals from fluorescence-based readout of the ensemble after the $\phi = 0$ and $\phi = \pi$ VPDR sequences.

A. Extraction of transition frequencies for each orientation

First, we examine whether we can accurately extract each orientation's $m_s = \pm 1$ transition frequency $\{\Delta\omega_i\}$ from a simulated dataset $S_{\text{VPDR}}(t_j, \tau_k)$ calculated at a discrete set of N pulse durations t_j and M free evolution times τ_k . For simplicity, we work with evenly spaced times along both dimensions.

The most straightforward inversion approach uses a frequency-domain analysis. Since the DQ term of interest has the form $\cos \Omega_{\text{eff}} t \cos \Delta\omega \tau$ [as can be found by

expanding powers in Eq. (2)], we consider a frequency-domain signal $I(\nu, \omega)$ obtained from a double-cosine inner product of S_{VPDR} given by

$$I(\nu, \omega) = \frac{\sum_k (f(\tau_k, \nu) - \bar{f}(\nu)) \cos \omega \tau_k}{\sum_k \cos^2 \omega \tau_k}, \quad (4)$$

$$f(\tau_k, \nu) = \frac{\sum_j S_{\text{VPDR}}(t_j, \tau_k) W(t_j) \cos \nu t_j}{\sum_j \cos^2 \nu t_j}, \quad (5)$$

where we subtract the mean value $\bar{f}(\nu) = 1/M \sum_k f(\tau_k, \nu)$ and allow different window functions $W(t_j)$. Figure 4(a) shows $I(\nu, \omega)$ for a boxcar window $W(t_j) = 1$, for a range of ν restricted to show only the $\nu = \Omega_{\text{eff}}$ features for each orientation; Ω_{eff} for the $\langle \bar{1}11 \rangle$ orientation is highlighted in white. The spectral peaks show fringing, particularly in the vertical dimension, which occurs because $S_{\text{VPDR}}(t, \tau)$ does not fully decay over the probed values of t_j and τ_k (especially in the pulse duration dimension t_j). These fringes pose a challenge because they lead to crosstalk between the different orientations' signals: the fringes from one orientation overlap the signal of another. We therefore switch to a Blackman window function [53] that suppresses the fringes at the cost of a slightly wider and shallower central lobe [see Fig. 4(a) right inset for the associated filter function]. With a Blackman window, we take a cut along the effective Rabi frequency (white line) to obtain the frequency-domain Ramsey signal $I(\sqrt{\Omega^2 + \omega^2}, \omega)$ for the $\langle \bar{1}11 \rangle$ orientation of interest [lower inset in Fig. 4(a)]. By fitting with a sum of Lorentzians, one might seek to estimate the $m_s = \pm 1$ transition frequency.

However, $I(\Omega_{\text{eff}}, \omega)$ is not exactly the sum of Lorentzians, even for ideal exponentially decaying sinusoidal signals in the time domain. Owing to the finite spacing in free evolution durations τ_k , there is a small deformation near zero frequency that distorts the fit and leads to systematic inaccuracies in the estimated transition frequencies [at the ~ 10 nT level in the example in Fig. 4(a)]. We thus consider the time-domain Ramsey signal $f(\tau_k, \nu = \Omega_i)$, where Ω_i is the Rabi frequency for the orientation of interest. Figure 4(b) shows $f(\tau_k, \Omega_{\langle \bar{1}11 \rangle})$ for the $\langle \bar{1}11 \rangle$ orientation with a Blackman window, fit to the sum of three exponentially decaying sinusoids. This time-domain analysis provides a significantly better estimate of the transition frequencies [54]. Comparing the extracted $\Delta\omega$ for the highest-frequency sinusoid with the exact transition frequency $\Delta\omega_{\text{exact}}$, we find the fit in Fig. 4(b) has an error of $\Delta\omega - \Delta\omega_{\text{exact}} \approx 20$ Hz, limited by residual crosstalk from the other orientations. By conversion to an equivalent axial magnetic field error $\Delta B = \hbar/2g\mu_B(\Delta\omega - \Delta\omega_{\text{exact}})$, we extract $\Delta B \approx 0.35$ nT.

While the time-domain inversion via fitting $f(\tau_k, \Omega_i)$ offers good accuracy, other approaches may prove superior depending on instrument goals. Acquiring data over a full

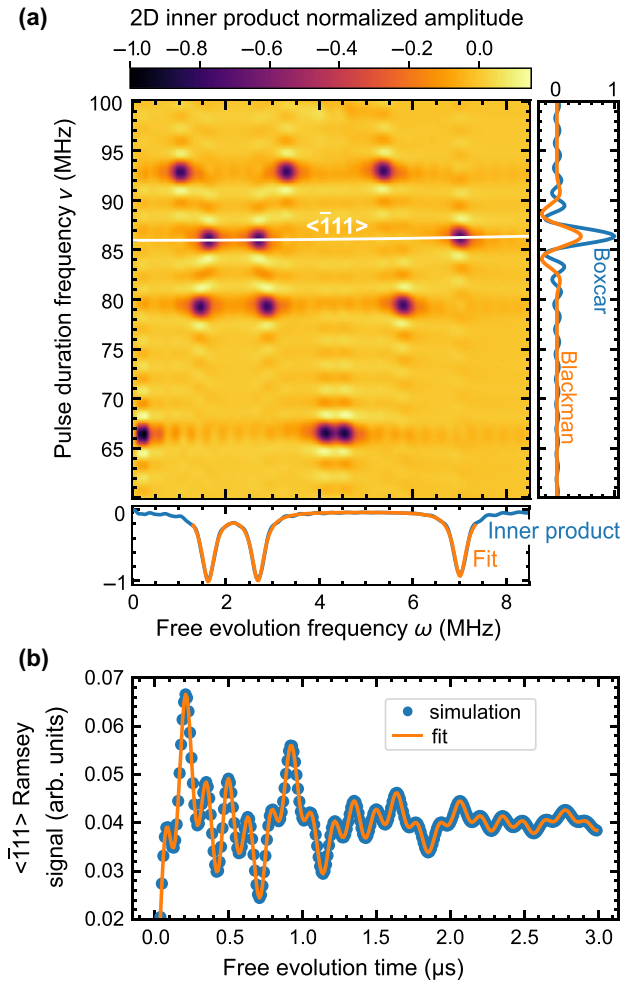


FIG. 4. Inner-product-based inversion. All plots in this figure analyze the same simulated single-quantum-canceled dataset $S_{\text{VPDR}}(t_j, \tau_k)$ performed for $\mathbf{B}_{\text{dc}} = (-38.4, 25.7, 19.1)$ μT in conventional diamond unit cell coordinates, at the optimum microwave angle from Fig. 3, with $\Omega_{\text{max}} = 100$ MHz, $T_2^* = 2$ μs , a maximum pulse duration t_j of 397.5 ns, and a pulse step size of 2.5 ns. (a) Boxcar-windowed two-dimensional inner product $I(\nu, \omega)$ as defined in Eqs. (4) and (5). Right inset: Filter functions $F(\nu) \propto \sum_j W(t_j) \cos \nu t_j$ for boxcar (blue) and Blackman (orange) windows. Lower inset: $I(\nu, \omega)$ evaluated along the white line in (a), with a Blackman window, fit with three Lorentzians. (b) Blackman-windowed τ -domain Ramsey signal $f(\tau_k) \propto \sum_j S_{\text{VPDR}}(t_j, \tau_k) W(t_j) \cos(\Omega_{\langle \bar{1}11 \rangle} t_j)$. The signal is fit with the sum of three exponentially decaying sinusoids with commensurate decay times, frequencies constrained to $\Delta\omega = 2|\omega_0 + m_l A|$ for $m_l \in \{-1, 0, 1\}$, and variable phase (due to an inner product at fixed Ω rather than Ω_{eff}).

set of MW pulse durations and free evolution times leads to sensitivity that is roughly 30 times worse than the sensitivity of single- τ DQ Ramsey measurements, depending on acquisition details (see Appendix B). Moreover, acquisition of the 48 000 measurements used for the example in Fig. 4 would require more than 1 s in typical systems, limiting bandwidth. Acquisition time could be reduced

by more than an order of magnitude by sampling only at free evolution times when all of the hyperfine signals rephase, $\tau_s = 2\pi s/(2A)$ for $s \in \mathbb{N}$; such sampling would improve sensitivity but limit dynamic range ($g\mu_B B_z/\hbar < A/2$). Bayesian adaptive schemes [55,56] as well as non-adaptive phase estimation algorithms [57,58] could also be applied in the context of the VPDR protocol.

In particular, if the goal is to sense small deviations from a known field, which is the context in which sensitivity is usually defined, one would measure only at the optimal free evolution time $\tau_{\text{opt}}^{(i)}$ for each orientation i . In this scenario, the cost to sensitivity of the VPDR protocol is minimal: the sensitivity for measuring a single orientation at $\tau_{\text{opt}}^{(i)}$ is only $2\sqrt{2}$ worse than that of standard DQ Ramsey measurement in the hard-pulse limit, and approximately 6 times worse for the conditions considered in Figs. 5 and 6 (see Appendix B). Furthermore, even though the evolution time is not optimal for them, some information is gained about other nontargeted orientations from VPDR signals, such that sequential VPDR measurements at all $\tau_{\text{opt}}^{(i)}$ hold some advantage over sequential DQ Ramsey measurements. Even in this scenario, the refresh rate is reduced by the number of pulse durations sampled, but it may be possible to devise sparse-sampling approaches in both t and τ dimensions. Here, we focus on the question of inversion accuracy and leave optimization of sensitivity and refresh rate to future development.

B. Inversion accuracy

We examine the accuracy of the time-domain inversion by simulating and analyzing VPDR signals over a range of magnetic fields. Figure 5 shows the systematic inversion error for a typical Earth magnetic field magnitude of 50 μT , expressed as the equivalent error ΔB in the axial field along each NV orientation. Subnanotesla accuracy is achieved for most dc field angles, with the $\langle 111 \rangle$ orientation showing the smallest systematic error because it enjoys the best spectral isolation from the other orientation's Rabi frequencies and harmonics. Away from the large-error contours discussed below, we observe detailed patterns in ΔB that vary with the field magnitude. If we increase the maximum Rabi pulse duration or eliminate nontarget orientations from the simulation, these errors are reduced by orders of magnitude, thus demonstrating that they are caused by crosstalk from other orientations.

Each orientation exhibits zones of poor accuracy when the dc field is perpendicular to its axis; the dashed-dotted line in the $\langle 1\bar{1}1 \rangle$ panel in Fig. 5 shows its perpendicular-field contour. These dead zones are associated with vanishing sensitivity of a cosine-like Ramsey signal at zero field, which is ultimately caused by the fixed direction of the applied MW field (see Appendix B 2 for further discussion). The loss of sensitivity exacerbates crosstalk effects and leads to poor accuracy. Fortunately, the dead

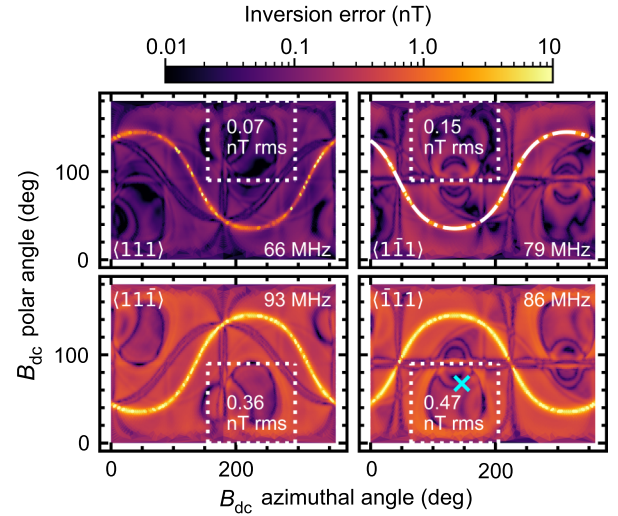


FIG. 5. Systematic inversion error for a $B_{\text{dc}} = 50 \mu\text{T}$ field with varying direction relative to diamond crystalline axes. Simulations are run with $\Omega_{\text{max}} = 100 \text{ MHz}$, $T_2^* = 2 \mu\text{s}$, microwave direction as shown in Fig. 3(b), pulse durations up to 797.5 ns in 2.5 ns steps, and free evolution times up to 2.98 μs in 20 ns steps. The time-domain Ramsey signal $f(\tau, \Omega)$ is calculated for each orientation with the use of a Blackman window and fit to extract the $m_s = \pm 1$ transition frequency of the highest-frequency hyperfine line. The error compared with the exact eigenvalue difference is expressed as the equivalent error in the axial field, with plots for each orientation labeled by their crystallographic direction and approximate Rabi frequency Ω . The dashed-dotted white line shows the region of vanishing axial field for the $\langle 1\bar{1}1 \rangle$ orientation. Dotted boxes indicate regions in which the indicated rms errors are calculated; the cyan cross marks the field and orientation for which simulations in Fig. 4 are calculated.

zone angular extent is small; furthermore, the dc field can be reconstructed from three orientations' signals, meaning that true dead zones are encountered only when the dc field is perpendicular to two NV axes simultaneously.

While we only show 50 μT data, we have also found similar performance in numerical simulations over terrestrial fields of 20–70 μT . The dynamic range is limited by the Nyquist frequency for sampling in τ , with the requirement that $2(\omega_L + A)/2\pi < 1/(2\Delta\tau)$ where $\Delta\tau = \tau_{k+1} - \tau_k$; for 20 ns sampling, the dynamic range exceeds 350 μT . Even with faster sampling, there will also be an upper limit on the range as ω_L approaches Ω_i : for $\Omega_i/\omega_L < 2$, there is a steady fall-off in sensitivity as the $|\pm\rangle$ states are too far detuned to be effectively driven [see Fig. 2(b)]. Since Ω_{max} cannot be significantly increased within the regime of validity for the rotating wave approximation, we can estimate an upper limit on the range of $\omega_L \sim 30 \text{ MHz}$ or $|B| \sim 1 \text{ mT}$.

C. Robustness regarding microwave drift

Up until this point, we have performed inversions assuming known MW amplitude and direction. However,

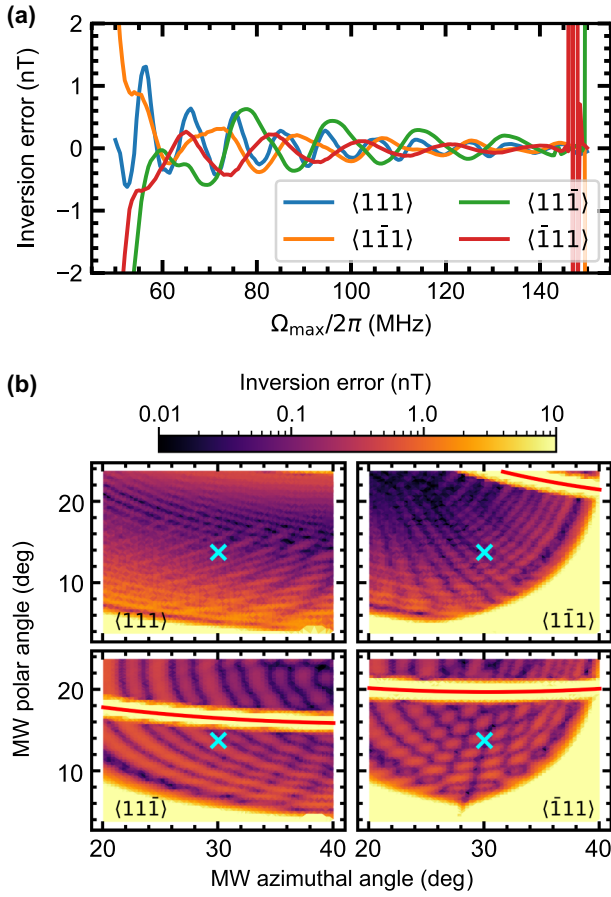


FIG. 6. Robustness regarding drift in microwave (MW) excitation parameters. (a) Inversion error vs microwave amplitude. (b) Inversion error vs microwave direction for $\Omega_{\max}/(2\pi) = 100$ MHz. Cyan crosses show the direction used in (a); red lines indicate coincidence of $3\Omega_{\langle 111 \rangle}/2$ with Ω_i . Yellow regions exceed 10 nT error. In (a),(b), simulations are run for $B_{dc} = (-38.4, 25.7, 19.1)$ μT (cyan cross in Fig. 5) with $T_2^* = 2$ μs , pulse durations up to 797.5 ns in 2.5 ns steps, and free evolution times up to 2.98 μs in steps of 20 ns. We perform inversions without prior knowledge of precise microwave parameters by estimating Ω_i from $\sum_k f(\tau_k, \nu)$ and then fitting $f(\tau_k, \Omega_i)$ for each orientation.

a primary strength of the VPDR protocol is that it does not require perfect calibration of the MW field; it is robust regarding slow drifts in MW amplitude and direction because the Rabi frequency for each orientation can be determined from the signal. In some sense, the labeling of NV orientations is self-calibrating. Qualitatively, by integrating $I(\nu, \omega)$ [see Fig. 4(a)] over ω , one obtains a Rabi spectrum whose features permit estimation of Ω_i . In practice, we calculate $\sum_k f(\tau_k, \nu)$, which exhibits peaks near $\nu = \Omega_i$. Provided the drifts in MW excitation are sufficiently small that the features retain their ordering, Ω_i can thereby be estimated for each NV axis. Furthermore, slightly imperfect Rabi-frequency estimation does not significantly impact the frequencies in the time-domain

Ramsey signal. Thus even fairly large drifts in the MW “label” do not cause concomitant systematic errors in the magnetic field measurement.

Figure 6 illustrates the robustness of the VPDR protocol regarding variation in MW power and direction when Rabi frequencies are estimated from simulated data. Inversions in Fig. 6 assume a known ordering of Rabi frequencies, corresponding to an approximate MW direction, but do not otherwise use *a priori* knowledge of MW parameters. As the MW amplitude increases, the inversion error ΔB decreases because the different orientations’ signals become better separated and crosstalk is reduced [see Fig. 6(a)]. At some point, the Rabi frequencies become so high that aliasing in the data becomes an issue, leading to the wild variations for $\Omega_{\max}/2\pi \gtrsim 145$ MHz; this could be mitigated by reducing the pulse duration step size $\Delta t = t_{j+1} - t_j$ in $S_{\text{VPDR}}(t_j, \tau_k)$. Regardless, Fig. 6(a) illustrates robustness regarding factor-of-2 variations in MW amplitude. Variations in MW direction [see Fig. 6(b)] produce acceptable errors over at least 8° ranges in the polar and azimuthal angles between the diamond crystalline axes and the MW field. The large range of tolerable MW angles and magnitudes seen in Fig. 6 is representative of typical results in terrestrial fields, though detailed crosstalk-induced features in the errors vary. In all cases, the angular limits are constrained by the $3\Omega_{\langle 111 \rangle}/2$ harmonic, which produces significant crosstalk as it approaches the other orientations’ Rabi frequencies (red lines).

The above analysis concerns quasistatic changes to the MW field, which are an essential consideration for long-term accuracy. MW intensity fluctuations on timescales faster than a measurement could elongate features along the pulse duration frequency axis, potentially increasing crosstalk [see Fig. 4(a)]. Nevertheless, some noise is tolerable provided that the associated Rabi broadening is small compared with the dephasing rate or the inverse of the maximum pulse duration. It is also worth noting that our analysis implies robustness regarding drifts in MW detuning due to, for example, temperature fluctuations. Detuning does not impact the double-quantum transition frequency, and the typical changes it induces in the effective Rabi frequency are tiny compared with the range that the VPDR protocol can tolerate.

Ultimately, these numerical investigations demonstrate the possibility to accurately extract NV spin transition frequencies along the four diamond bond directions, without relying on precision calibration of the MW direction used to label the NV orientations.

IV. EXPERIMENTAL FEASIBILITY STUDY

To probe the experimental feasibility of realizing the VPDR protocol and verify its qualitative features, we performed preliminary tests with an improvised device. Figure 7(a) illustrates the key capabilities of the system, which

we constructed by adapting an existing continuous-wave ODMR magnetometer prototype. The diamond sample is mounted on a rod within a three-dimensional (3D) MW resonator [59] between two bow-tie pillars that determine the direction of the MW field, while permitting relatively high Rabi frequencies in the tens of megahertz range. Rotation of the rod enables partial control of the orientation of the diamond crystal axes relative to the driving MW field. NV centers in the sample are illuminated with a 520 nm laser (approximately 100 mW), and fluorescence is detected by a photodiode below the diamond. MW pulses are generated with a voltage-controlled oscillator gated by a field-programmable-gate-array-based (FPGA-based) controller. The entire device can be placed inside a uniaxial solenoid coil within high-permeability shielding to explore variation with the external magnetic field.

As the diamond is rotated relative to the MW resonator, the Rabi frequencies for each orientation vary. Figure 7(b) shows the fast Fourier transform (FFT) of the ensemble Rabi oscillation as a function of the rotation angle, clearly illustrating four distinct Rabi frequencies (split by hyperfine interactions, particularly at lower Rabi frequency). While we can reach angles that adequately separate the bare Rabi frequencies Ω_i of the four NV orientations, the ideal angle shown in Fig. 3(b) is not attainable, and some of the Rabi harmonics overlap. Furthermore, in this prototype we do not have *a priori* knowledge of the absolute diamond orientation, so we cannot assign Rabi frequencies to specific crystallographic directions.

At a diamond rotation angle of approximately 73° , we perform the VPDR protocol as a function of the voltage sent to the uniaxial coil to verify that anticipated features are observable and move with the field as expected. Because the MW delivery system did not have dynamic phase control capabilities, we identify the DQ signals by averaging data over detuning. Unlike DQ signals, SQ signals occur at free evolution frequencies that change with detuning, so the detuning-averaged SQ signals are smeared out horizontally in the Fourier domain, whereas the DQ signals remain pointlike.

Figure 7(c) shows a sample Fourier-domain dataset obtained from an average over three detunings at a solenoid coil voltage of 4 V. The Fourier representation uses a mean-subtracted double inner product given by Eqs. (4) and (5) with complex exponentials replacing the cosines [60], using a cosine window function. Local peaks in the Fourier-domain VPDR data at large free-evolution frequencies can be attributed to DQ signals from each of the four NV orientations. Crosses mark local extrema associated with an $m_I = +1$, DQ feature from each orientation. We find a local maximum in the inner product magnitude to obtain the extrema locations $(\omega_{\max,i}, \nu_{\max,i})$ along with their inferred harmonic n (where $\nu_{\max,i} = n\Omega_{\text{eff},i}$), and thereby extract the

bare Rabi frequency $\Omega_i = \sqrt{\nu_{\max,i}^2/n^2 - \omega_{\max,i}^2}$, approximate field projection $|B_{z,i}| = \hbar|\omega_{\max,i} - 2A|/(2g\mu_B)$, and expected locations of other DQ features from the same orientation (marked with dots of the same color). While the agreement is not perfect, we do observe features at or near many of the expected locations, particularly for the orientations marked in green, blue, and orange. The poor visibility of the fourth orientation (red) could be explained by optical polarization selection rules. The numerous peaks at moderate free evolution frequency are attributed to SQ signals, which are horizontally tripled owing to the three detunings over which we average the data.

We perform the analysis illustrated in Fig. 7(c) for a range of solenoid coil voltages, extracting the Rabi frequency [Fig. 7(d)] and axial field projection [Fig. 7(e)] for each orientation. The Rabi frequencies Ω_i remain relatively constant with voltage, and occur at values compatible with the Rabi FFT data considering that several days elapsed between the Rabi and VPDR data acquisitions [see Fig. 7(b), dotted white line]. In contrast, the field projections evolve significantly with the coil voltage. To determine whether our observations are consistent with a uniaxial field that increases linearly with the coil voltage, we fit our data to field projections from a linear model [see Fig. 7(f) and Appendix D]. With no absolute knowledge of the diamond orientation in our resonator, we could not unambiguously match the ordering of Rabi frequencies to an ordering of known NV orientations, although such identification could be achieved by calibration in triaxial coils [61,62]. We also do not know the sign of the field projection on each NV axis (which could be determined by pulsing a small magnetic field with approximately known direction). As a consequence, the inversion in Figs. 7(e) and 7(f) is not unique (different permutations and signs of the field projections can yield the same result), but the good correspondence between the fit and the data indicates that our measurements follow the expected linear behavior and that the projection on the fourth orientation is compatible with the projections on the other three orientations. The offset in the field at zero coil voltage can be explained by magnetic components in the rotation stage holding the diamond.

While the qualitative features of the data behave as expected, we leave quantitative tests of accuracy to future experiments. In Fig. 7(e), we observe rms deviations of $0.7 \mu\text{T}$ between the data and the fit, but these likely arise from nonidealities of our apparatus. The coil was not calibrated, and the voltage source used has an accuracy of $0.05\% + 10 \text{ mV}$, corresponding to $0.3\text{--}0.4 \mu\text{T}$ over the field range inferred from the fit. Furthermore, the magnetometer used for these tests presented additional sources of systematic error. The diamond support rod blocked the access path for detecting transmitted laser light, which normally would be used as a reference to account for

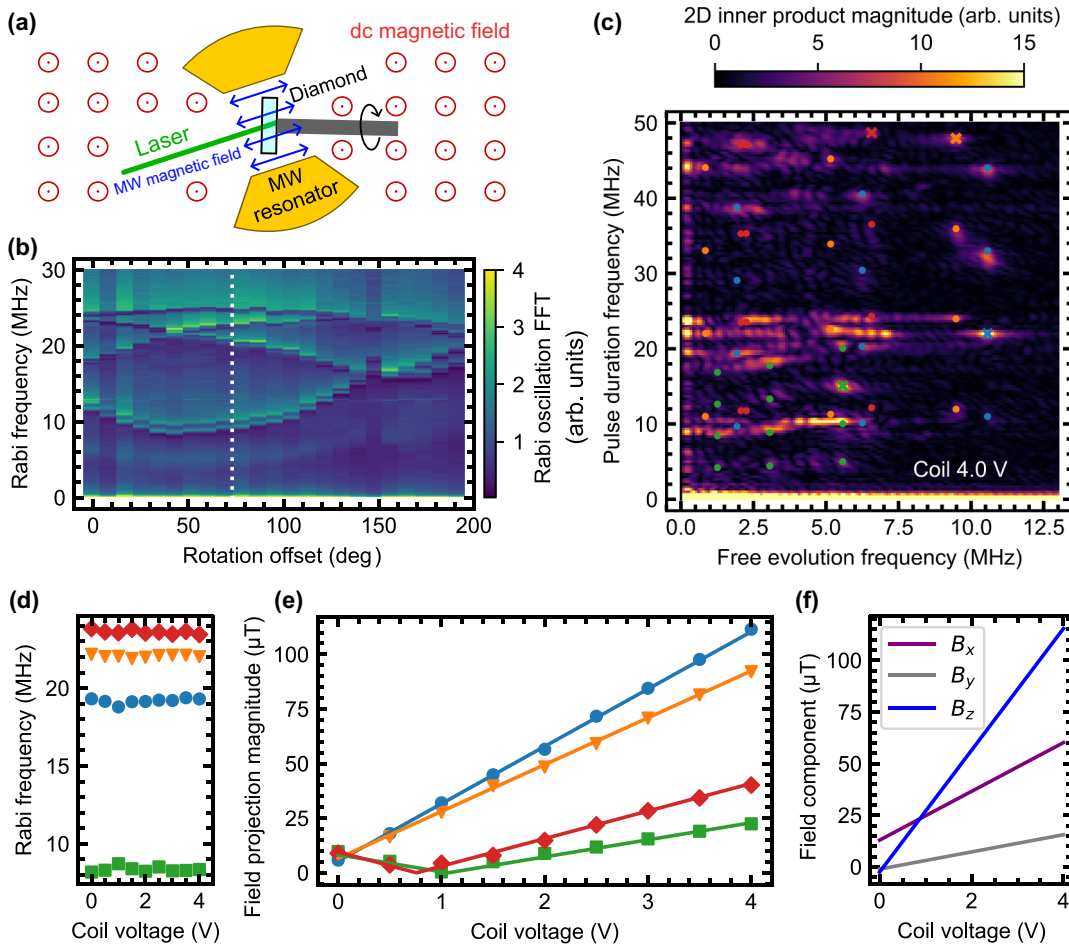


FIG. 7. Experimental feasibility study. (a) Top view of the device illustrating the ability to rotate the diamond relative to the microwave (MW) magnetic field. (b) Fast Fourier transform (FFT) of Rabi oscillation data as a function of diamond angle in the absence of an applied dc field. The angle chosen for experiments is indicated by the dashed white line. (c) Example Fourier-domain dataset at a coil voltage of 4 V. Variable-pulse-duration Ramsey data are acquired at pulse durations from 50 ns to 1.995 μs in 5 ns steps and free evolution times from 50 ns to 2.99 μs in 30 ns steps, and are averaged over acquisitions at microwave frequencies of 2.867, 2.8674, and 2.8678 GHz. Local maxima associated with a double-quantum signal are marked with a cross and used to infer expected locations of other double-quantum peaks for the same orientation, indicated by dots of the same color. (d) Inferred bare Rabi frequency found from inner-product maxima [crosses in (c)] as a function of coil voltage. Different symbols are associated with distinct but unknown NV orientations. (e) Inferred projections of the magnetic field on each orientation's axis. Solid lines show field projection magnitudes from a fit to a magnetic field that varies linearly with voltage. (f) Field components, expressed in crystal coordinates, of the linearly varying field fit to the inferred field projections in (e). Note that this field is not unique.

fluctuations in laser power; there was also observable electronic crosstalk, mechanical vibration, and thermal drift. These issues added significant systematic uncertainty, but they are straightforward to remedy. Similarly, adding a phase-control circuit would suppress the SQ features that complicate the analysis presented. Nevertheless, even with poor data quality, it was possible to interpret VPDR features and extract magnetic field estimates that are consistent with the linearly increasing applied field, providing a proof-of-principle demonstration of the VPDR protocol.

One notable shortcoming of our device was its microwave resonator, which was not purpose-designed and could reach only ~ 30 MHz Rabi frequency. Future devices

could be optimized to increase both MW strength and MW homogeneity. While planar resonators have reached 165 MHz Rabi frequencies [63] and coil-based MW antenna designs can exhibit 60 MHz Rabi frequency over a homogeneous volume of order $\sim 1 \text{ mm}^3$ [64], 3D re-entrant cavities with bow-tie posts [59] offer excellent homogeneity and spatial field confinement, and we anticipate that careful optimization of post geometry [65] will permit 100 MHz to be reached. Indeed, the fundamental limit on the Rabi frequency set by the resonator Q , frequency f , effective volume V , and input power P_{in} permits Rabi frequencies of ~ 330 MHz at $Q = 100$, $V = (1 \text{ mm})^3$, and $P_{\text{in}} = 10 \text{ W}$ [66]. Additionally, our simulations show subnanotesla rms

errors for $\Omega_{\max}$ down to 60 MHz. Taken together, these considerations show the feasibility of future quantitative experimental tests of VPDR accuracy.

V. CONCLUSION

The VPDR protocol offers a new paradigm for bias-field-free vector NV magnetometry, using Rabi labeling to isolate the signal from different orientations in a NV ensemble with minimal crosstalk. Our analytic modeling provides a resource for understanding and interpreting VPDR data, as well as identifying optimal regimes of operation. Numerical simulations of the protocol demonstrate its compatibility with high-accuracy field estimation, even when the MW field used to label NV orientations is only approximately known. Practically, the scheme requires only a single MW frequency, and is not sensitive to slow changes in detuning. The primary design requirement for experimental implementation is a relatively strong and homogeneous MW drive field, and VPDR data could be measured and interpreted even with an improvised device. The protocol avoids the use of additional hardware, and permits bias-field-free vector field measurements in a compact geometry.

There remain some challenges to working with the VPDR protocol instead of a bias field. A mechanism to determine the sign of the magnetic field projection on each NV axis is still required, e.g., by pulsing a coil to add a known magnetic field shift and observe how each orientation's signal changes. Note that the direction and magnitude of such a pulsed field need only be known approximately to obtain the sign information. In addition, sensitivity dead zones will occur where the external field is perpendicular to two NV axes, requiring multiple sensors or piezoelectric actuation, similar to designs for atomic magnetometers [67,68]. Finally, the simple data acquisition and inversion proposed here precludes high-bandwidth sensing, although it is likely that faster approaches exist.

Indeed, there are opportunities for refinement of the VPDR protocol as well as its numerical modeling and experimental implementation. MW resonator design targeting power and homogeneity would greatly simplify data interpretation and inversion. More sophisticated theory including internal electric fields [69,70] and nonsecular hyperfine terms [71] may be required for high-accuracy interpretation of experimental data in low fields. Increased bandwidth and improved sensitivity can likely be achieved with different sampling approaches and inversion methods, and it may be advantageous to incorporate existing sensitivity-enhancement techniques such as P1 driving [46]. It may also be possible to adapt our approach to other defect magnetometer technologies such as the V_{Si} in SiC [72]. With a recent surge of interest in low-field [27,73,74] and high-accuracy [75,76] NV magnetometry, the VPDR

protocol provides a timely opportunity to eliminate bias fields and improve the accuracy of NV vector-field sensors.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [77].

APPENDIX A: ANALYTIC MODEL AT FINITE Ω/ω_L AND WITH DEPHASING

The probability for an $m_s = 0$ -prepared spin to end up in $m_s = 0$ after application of the VPDR pulse sequence is given by $P_0(t, \tau)$. For a coherently evolving single NV center in an axial magnetic field, we can analytically calculate this probability by solving the Schrödinger equation with a piecewise-constant Hamiltonian given by H_1 from time $x = 0$ to $x = t$, H_0 from $x = t$ to $x = t + \tau$, and $H_{1\phi}$ from $x = t + \tau$ to $x = 2t + \tau$. H_1, H_0 , and $H_{1\phi}$ are derived from H given in Eq. (1) as follows: In the $\Omega \rightarrow \infty$ limit, we set $H_1 = H(\phi = 0, \delta = 0, \omega_L = 0)$, $H_0 = H(\Omega = 0)$, and $H_{1\phi} = H(\delta = 0, \omega_L = 0)$, for which simple analytic solutions can be found. For finite- Ω/ω_L calculations, we set $H_1 = H(\phi = 0, \delta = 0)$, $H_0 = H(\Omega = 0, \delta = 0)$, and $H_{1\phi} = H(\delta = 0)$, allowing for finite ω_L during all time segments; we restrict our calculations to zero detuning because it keeps the expressions relatively simple and reflects conditions under which the protocol is likely to be run.

At finite Ω/ω_L and zero detuning $\delta = 0$, the VPDR signal for a single NV center without hyperfine structure comprises oscillations at $v_n = n\sqrt{\Omega^2 + 4\omega_L^2} = n\Omega_{\text{eff}}$ and $\omega_m = m\omega_L$ in t and τ , respectively, where $n = \{-2, -\frac{3}{2}, -1, -\frac{1}{2}, 0, \frac{1}{2}, 1, \frac{3}{2}, 2\}$ and $m = \{-2, -1, 0, 1, 2\}$.

In the absence of dephasing, the $m_s = 0$ population after the VPDR pulse sequence can be found analytically and expressed as

$$P_0(t, \tau) = \sum_n \sum_m a_{n,m} e^{i\nu_n t} e^{i\omega_m \tau}. \quad (\text{A1})$$

The coefficients $a_{n,m}$ depend only on the ratio $\alpha = \Omega/\omega_L$ and the relative phase ϕ of the second MW pulse, and $a_{n,m}$ are given in Table I for $n \geq 0$. Since $P_0(t, \tau)$ is real-valued, the remaining coefficients can be found from the relationship $a_{-n,-m} = a_{n,m}$.

To include dephasing in the model in an analytically tractable manner, we ignore spin dephasing during the pulses because we expect $\max(t) \ll \max(\tau)$ in experiments using high Rabi frequency $\Omega \gg 1/T_2^*$. We also assume that the spin dephasing is quasistatic and magnetic in nature, and we model it using a random internal axial magnetic field B_z^0 parameterized by a Lorentzian probability distribution of the associated Larmor frequencies $g(\omega_L^0) = T_2^*/\pi ((T_2^*\omega_L^0)^2 + 1)$ [47]. In this quasistatic model, each spin evolves according to Eq. (A1) with a Larmor precession frequency $\omega_L + \omega_L^0$, with ω_L^0 randomly chosen from the distribution $g(\omega_L^0)$, such that the total average probability of observing the spins in $m_s = 0$ is the convolution of $P_0(t, \tau)$ (as a function of ω_L) and g . The average signal is thereby given by

$$\begin{aligned} \bar{P}_0(t, \tau) &= \sum_n \sum_m a_{n,m} e^{i\nu_n t} \int_{-\infty}^{\infty} g(x - \omega_L) e^{i\omega_m \tau} dx \\ &= \sum_n \sum_m a_{n,m} e^{-|m|\tau/T_2^*} e^{i\nu_n t} e^{i\omega_m \tau}, \end{aligned} \quad (\text{A2})$$

where n and m are summed over the ranges given for Eq. (A1). Here we have ignored the impact of ω_L^0 on Ω_{eff} , which is consistent with our ignoring dephasing during the pulses and is an approximation appropriate for $\Omega \gg \omega_L, 1/T_2^*$.

Note that even in the finite- Ω/ω_L regime with dephasing, the coefficients of the SQ terms (corresponding to $m = \pm 1$) are proportional to $\cos \phi$, indicating that the SQ terms can be eliminated by addition of two signals acquired with (1) $\phi = 0^\circ$ and (2) $\phi = 180^\circ$ relative phases for the MW pulses.

APPENDIX B: SENSITIVITY ANALYSIS

1. Comparison with standard double-quantum Ramsey measurements

a. Hard-pulse limit $\Omega/\omega_L \rightarrow \infty$

Here, we analyze the sensitivity cost of the VPDR protocol relative to a more typical double-quantum Ramsey experiment performed at fixed pulse duration $t = \pi/\Omega$ in near-zero field. For simplicity, we imagine a situation

TABLE I. The coefficients $a_{n,m}$ for $P_0(t, \tau) = \sum_n \sum_m a_{n,m} e^{i\nu_n t} e^{i\omega_m \tau}$ for a single NV center without hyperfine structure at zero detuning and with no dephasing. Expressions are given in terms of $\alpha = \Omega/\omega_L$ and $\tilde{\alpha} = \sqrt{\alpha^2 + 4}$ for concision.

n	$m = -2$	$m = -1$	$m = 0$	$m = 1$	$m = 2$
2	$\frac{\alpha^8(\tilde{\alpha}-8)+32\alpha^6(\tilde{\alpha}-3)+128\alpha^4(\tilde{\alpha}-2)}{64\tilde{\alpha}^9}$	$\frac{\alpha^6(\alpha^2(\tilde{\alpha}-4)+8(\tilde{\alpha}-2))\cos(\phi)}{16\tilde{\alpha}^9}$	$\frac{3\alpha^8}{32\tilde{\alpha}^8}$	$\frac{\alpha^6(\alpha^2(\tilde{\alpha}+4)+8(\tilde{\alpha}+2))\cos(\phi)}{16\tilde{\alpha}^9}$	$\frac{\alpha^8(\tilde{\alpha}+8)+32\alpha^6(\tilde{\alpha}+3)+128\alpha^4(\tilde{\alpha}+2)}{64\tilde{\alpha}^9}$
$\frac{3}{2}$	$\frac{\alpha^6(20-6\tilde{\alpha})-32\alpha^4(\tilde{\alpha}-2)+\alpha^8}{4\tilde{\alpha}^9}$	$\frac{\alpha^4(16(\tilde{\alpha}-2)+\alpha^4-4\alpha^2)\cos(\phi)}{2\tilde{\alpha}^9}$	$\frac{3\alpha^6}{\tilde{\alpha}^8}$	$\frac{\alpha^4(16(\tilde{\alpha}+2)-\alpha^4+4\alpha^2)\cos(\phi)}{2\tilde{\alpha}^9}$	$\frac{\alpha^4(\alpha^2(6\tilde{\alpha}+20)+32(\tilde{\alpha}+2)+\alpha^4)}{4\tilde{\alpha}^9}$
1	$-\frac{\alpha^4(\alpha^2-24)(\alpha^2(\tilde{\alpha}-4)+8(\tilde{\alpha}-2))}{16\tilde{\alpha}^9}$	$-\frac{\alpha^2(-4\alpha^4(\tilde{\alpha}+3)+8\alpha^2(3\tilde{\alpha}-4)-64(\tilde{\alpha}-2)+\alpha^6)\cos(\phi)}{2\tilde{\alpha}^9}$	$\frac{\alpha^4(\alpha^4-16\alpha^2+256)}{8\tilde{\alpha}^8}$	$\frac{\alpha^2(4\alpha^4(\tilde{\alpha}-3)-8\alpha^2(3\tilde{\alpha}+4)+64(\tilde{\alpha}+2)+\alpha^6)\cos(\phi)}{2\tilde{\alpha}^9}$	$-\frac{\alpha^4(\alpha^2-24)(\alpha^2(\tilde{\alpha}+4)+8(\tilde{\alpha}+2))}{16\tilde{\alpha}^9}$
$\frac{1}{2}$	$-\frac{\alpha^4(3\alpha^2-16)(-2\tilde{\alpha}+\alpha^2+4)}{4\tilde{\alpha}^9}$	$\frac{\alpha^2(-4\alpha^4(2\tilde{\alpha}+1)+16\alpha^2(3\tilde{\alpha}+2)-128(\tilde{\alpha}-2)+\alpha^6)\cos(\phi)}{2\tilde{\alpha}^9}$	$\frac{\alpha^2(5\alpha^4-32\alpha^2+128)}{\tilde{\alpha}^8}$	$\frac{\alpha^2(\alpha^4(8\tilde{\alpha}-4)+\alpha^2(32-48\tilde{\alpha}+128(\tilde{\alpha}+2)+\alpha^6)\cos(\phi)}{2\tilde{\alpha}^9}$	$\frac{\alpha^4(3\alpha^2-16)(2\tilde{\alpha}+\alpha^2+4)}{4\tilde{\alpha}^9}$
0	$\frac{\alpha^4(3\alpha^4-96\alpha^2+128)}{32\tilde{\alpha}^8}$	$-\frac{\alpha^2(\alpha^6-24\alpha^4+320\alpha^2-512)\cos(\phi)}{8\tilde{\alpha}^8}$	$\frac{9\alpha^8+64\alpha^6+1536\alpha^4+4096}{16\tilde{\alpha}^8}$	$-\frac{\alpha^2(\alpha^6-24\alpha^4+320\alpha^2-512)\cos(\phi)}{8\tilde{\alpha}^8}$	$\frac{\alpha^4(3\alpha^4-96\alpha^2+128)}{32\tilde{\alpha}^8}$

where there is only a single NV orientation since low-field “standard” DQ Ramsey measurements would work only in this scenario. We assume that we are operating in the $\Omega/\omega_L \rightarrow \infty$ limit such that simple analytic expressions apply, and we assume the readout is characterized by Gaussian noise independent of the readout value (as appropriate for low-contrast fluorescence readout). Since sensitivity is usually defined in the context of a known initial field (from which one senses a small change), we consider measurements at a single free evolution time τ that is the same for the DQ Ramsey and VPDR schemes.

For the VPDR measurement, we consider an experiment comprising $N \gg 1$ measurements of $P_0(t_j, \tau)$ at pulse durations $t_0, \dots, t_{N-1}$, producing a fluorescence signal $S = a + bP_0(t_j, \tau)$, where a and b depend on the details of the measurement scheme. The signal is analyzed via an inner product with $\cos \Omega t_j$, possibly with a windowing function $W(t_j)$ applied. This procedure defines the analyzed signal

$$f_{\text{VPDR}}(\tau) = \frac{\sum_{j=0}^{N-1} W(t_j) \cos \Omega t_j (a + bP_0(t_j, \tau))}{\sum_{j=0}^{N-1} \cos^2 \Omega t_j}, \quad (\text{B1})$$

whose uncertainty can be found via standard Gaussian uncertainty propagation as

$$\Delta f_{\text{VPDR}} = \Delta S \frac{\sqrt{\sum_{j=0}^{N-1} W^2(t_j) \cos^2 \Omega t_j}}{\sum_{j=0}^{N-1} \cos^2 \Omega t_j}, \quad (\text{B2})$$

where ΔS is the uncertainty in each fluorescence measurement (assumed constant as appropriate for low contrast). In the limit that the t_j are evenly spaced over a range much larger than $2\pi/\Omega$ with a spacing much smaller than $2\pi/\Omega$, these sums can be approximated by integrals, such that $\sum_{j=0}^{N-1} \cos^2 \Omega t_j \rightarrow N/2$. If we can also assume that $W(t)$ varies slowly compared with the oscillations at frequency Ω , then in the numerator we can divide the sum up into M chunks where $W(t)$ is approximately constant during each chunk, and the sinusoidal signals can still be approximated by their average values integrated over many oscillations. In particular, if $t_j = j \Delta t$ and M evenly divides N , this chunking would look like

$$\begin{aligned} & \sum_{j=0}^{N-1} W(t_j) \cos \Omega t_j (a + bP_0(t_j, \tau)) \\ & \approx \sum_{j=0}^{M-1} W(t_{j \frac{N}{M}}) \underbrace{\sum_{i=0}^{N/M-1} \cos \Omega t_{j \frac{N}{M} + i} (a + bP_0(t_{j \frac{N}{M} + i}, \tau))}_{\left(\frac{N}{M}\right) \frac{b}{4} \sin^2 \omega_L \tau} \end{aligned}$$

where we have used

$$\lim_{T \rightarrow \infty} \frac{1}{2T} \int_{-T}^T dt \cos \Omega t P_0(t, \tau) = \frac{1}{4} \sin^2 \omega_L \tau \quad (\text{B3})$$

(in the $\Omega/\omega_L \rightarrow \infty$ limit) to approximate the inner sum. Since the inner sum no longer depends on j , we can approximate the outer sum by $M \bar{W}$, where $\bar{W}$ is the mean value of the windowing function. Ultimately, including the factor of $2/N$ from the denominator of Eq. (B1), we obtain

$$f_{\text{VPDR}}(\tau) \approx \frac{b}{2} \bar{W} \sin^2 \omega_L \tau, \quad (\text{B4})$$

and, by similar reasoning,

$$\Delta f_{\text{VPDR}} \approx \Delta S \sqrt{\frac{2}{N}} \sqrt{\bar{W}^2}. \quad (\text{B5})$$

We compare this analyzed VPDR signal with the signal obtained from averaging N repetitions of a standard double-quantum Ramsey experiment,

$$f_R = a + b \cos^2 \omega_L \tau, \quad (\text{B6})$$

$$\Delta f_R = \Delta S \frac{1}{\sqrt{N}}. \quad (\text{B7})$$

The sensitivity of each experiment is given by

$$\eta = \Delta B \sqrt{T} = \frac{\hbar}{g\mu_B} \Delta \omega_L \sqrt{T} = \frac{\hbar}{g\mu_B} \frac{\Delta f}{|df/d\omega_L|} \sqrt{T}, \quad (\text{B8})$$

where T is the total time required for an experiment that results in magnetic field uncertainty ΔB . For $\Omega \gg \omega_L$, the time required for the Rabi pulses is negligible compared with the free evolution and measurement time, so we can approximate that T is the same for both protocols. Thus, we can compare the sensitivity of the VPDR and double-quantum Ramsey experiments by finding the ratio of their signal to noise:

$$\frac{\eta_{\text{VPDR}}}{\eta_R} = \frac{\Delta f_{\text{VPDR}}}{|df_{\text{VPDR}}/d\omega_L|} \frac{|df_R/d\omega_L|}{\Delta f_R} \quad (\text{B9})$$

$$\approx \frac{\Delta S \sqrt{\bar{W}^2} \sqrt{2/N}}{b \bar{W} \tau |\sin 2\omega_L \tau|/2} \frac{b \tau |\sin 2\omega_L \tau|}{\Delta S / \sqrt{N}} \quad (\text{B10})$$

$$\approx 2\sqrt{2} \frac{\sqrt{\bar{W}^2}}{\bar{W}}. \quad (\text{B11})$$

If no windowing function is needed, we attain the best VPDR sensitivity at $2\sqrt{2}$ times the standard double-quantum Ramsey sensitivity.

Equation (B11) is derived in the $\Omega/\omega_L \rightarrow \infty$ limit, and as the weight of the relevant VPDR Fourier peaks varies

with Ω/ω_L , so too will the sensitivity. If we consider finite Ω at $\delta = 0$, with SQ subtraction, analyzed by an inner product with $\cos \Omega_{\text{eff}}$, the only modification to the above analysis is a reduced weight of the inner-product-extracted DQ Ramsey signal, yielding

$$\frac{\eta_{\text{VPDR}}}{\eta_R} \approx 2\sqrt{2} \frac{\sqrt{\overline{W}^2}}{\overline{W}} \frac{(\alpha^2 + 4)^4}{|\alpha^4 (\alpha^4 - 16\alpha^2 - 192)|}, \quad (\text{B12})$$

where $\alpha = \Omega/\omega_L$. Unsurprisingly, Eq. (B12) is well approximated by Eq. (B11) for $\Omega/\omega_L \gtrsim 10$.

b. Numerical simulations of sensitivity

Equation (B11) holds in the presence of dephasing during free evolution, but the sensitivity is also affected by dephasing during the MW pulses. To elucidate this effect, we performed Monte Carlo estimates of the $\eta_{\text{VPDR}}/\eta_R$ ratio as a function of the maximum MW pulse duration used in the VPDR protocol, assuming that the VPDR and DQ Ramsey experiments were both performed at the same optimal free evolution time τ_{opt} . Specifically, we simulated the SQ-canceled signal $S_{\text{VPDR}}(t_j, \tau_{\text{opt}})$ for a single NV orientation (such that standard low-field DQ Ramsey measurements will also work) both with and without hyperfine structure, and extracted $f_{\text{VPDR}}(\tau_{\text{opt}})$ [via Eq. (5)] and $f_R(\tau_{\text{opt}}) = S_{\text{VPDR}}(t_\pi, \tau_{\text{opt}})$. We calculated the slopes $df/d\omega_L$ for each by performing the simulation at slightly different magnetic fields (differing in magnitude by 1 nT). Then, we added randomly generated Gaussian noise with standard deviation $\sigma = 10^{-4}$ to $S_{\text{VPDR}}(t, \tau_{\text{opt}})$ (σ corresponds to few-nanotesla field excursions, well within the linear regime), from which we obtain a noisy value of $f_{\text{VPDR}}(\tau_{\text{opt}})$ via Eq. (5). The uncertainty Δf_{VPDR} is estimated by the standard deviation of our results. The corresponding uncertainty in $f_R(\tau_{\text{opt}})$ is $\sigma/\sqrt{N}$, where N is the number of MW pulse durations used to calculate $f_{\text{VPDR}}(\tau_{\text{opt}})$, effectively averaging over N noisy measurements of $S_{\text{VPDR}}(t_\pi, \tau_{\text{opt}})$. The uncertainty in $f_{\text{VPDR}}(\tau_{\text{opt}})$ and $f_R(\tau_{\text{opt}})$ is converted to uncertainty in the magnetic field by dividing it by the respective slope $df/d\omega_L$, yielding ΔB_p^{opt} for each protocol $p \in \{\text{VPDR}, \text{R}\}$. The resulting ratio of magnetic field uncertainties $\Delta B_{\text{VPDR}}^{\text{opt}}/\Delta B_R^{\text{opt}}$ is a good approximation to the axial field sensitivity ratio $\eta_{\text{VPDR}}(\tau_{\text{opt}})/\eta_R(\tau_{\text{opt}})$ in the limit that the spin initialization, readout, and free evolution take significantly more time than the MW pulses (which is well satisfied in typical ensemble experiments).

As an example [see Fig. 8(a)], we show Monte Carlo estimates of $\eta_{\text{VPDR}}(\tau_{\text{opt}})/\eta_R(\tau_{\text{opt}})$ for the $\langle 1\bar{1}1 \rangle$ orientation with $T_2^* = 2 \mu\text{s}$, detuning $\delta = 0$, the same dc magnetic field as for Figs. 4 and 6, and the same MW field as for Figs. 4 and 5; the $\langle 1\bar{1}1 \rangle$ orientation's 79.2 MHz Rabi frequency yields a π pulse of 6.32 ns, and we used $\tau_{\text{opt}} = 862$ ns for $m_I = 0$ only and $\tau_{\text{opt}} = 1.157 \mu\text{s}$ when all three

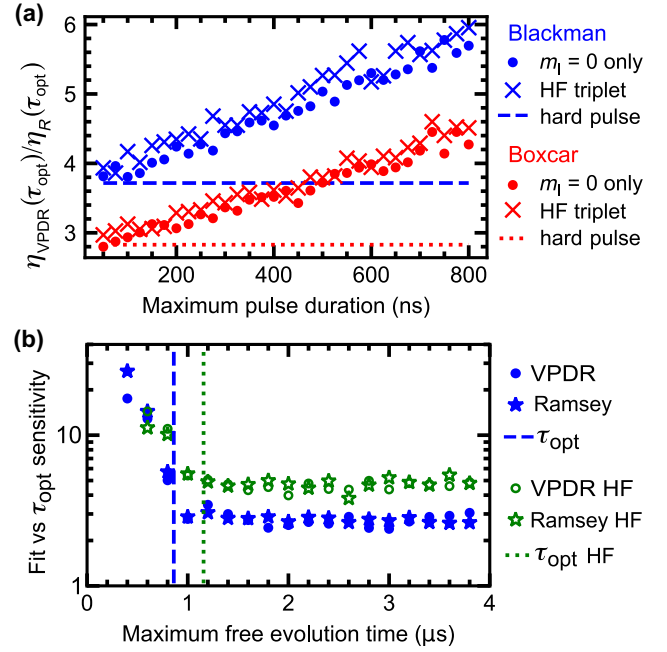


FIG. 8. Sensitivity cost associated with multiple microwave pulse durations and multiple free evolution times. Simulations are performed for the same magnetic field as in Fig. 6, and a microwave drive as in Fig. 5, considering solely the $\langle 1\bar{1}1 \rangle$ orientation. See the text for additional details. (a) Sensitivity cost of many microwave durations. Monte Carlo estimates of $\eta_{\text{VPDR}}(\tau)/\eta_R(\tau)$ for $\tau = \tau_{\text{opt}}$ are shown as a function of the maximum pulse duration used in the variable-pulse-duration Ramsey (VPDR) scheme, both with (blue) and without (red) a Blackman window on the pulse duration dimension, and both with (crosses) and without (circles) hyperfine (HF) structure. Dashed and dotted lines show the hard-pulse limit with and without a Blackman window, respectively. (b) Sensitivity cost of many free evolution times. For each protocol, Monte Carlo simulations estimate the ratio of the sensitivity from fitting signals $f_p(\tau_1, \tau_2, \dots)$ obtained at free evolution times $\tau_k < \tau_{\text{max}}$ to the sensitivity at the optimal free evolution time $\eta_p(\tau_{\text{opt}})$, where $p \in \{\text{VPDR}, \text{R}\}$. Evolution both with (green) and without (blue) hyperfine structure is considered. Dashed (dotted) lines show τ_{opt} in the absence (presence) of hyperfine structure.

hyperfine lines are included (both τ_{opt} values are found from analytic expressions for the ideal Ramsey signal in the hard-pulse limit; numerical optimization of simulated slopes yielded similar sensitivity ratios). For the VPDR scheme, the pulse durations t_j were evenly spaced by 2.5 ns, and included all t_j strictly less than the maximum pulse duration on the x axis of Fig. 8(a). This analysis was repeated for both boxcar and Blackman window functions.

Figure 8(a) quantifies the sensitivity cost of sampling at many MW pulse durations in the presence of dephasing. While the sensitivity ratio approaches the hard-pulse limit $2\sqrt{2}\sqrt{\overline{W}^2}/\overline{W}$ when the pulse durations are much shorter than T_2^* , for the long pulse durations up to 800 ns included in the analysis of Figs. 5 and 6, we observe a

sensitivity cost that is roughly 50% higher. Similar results are observed with and without hyperfine structure; we also see similar results for different NV orientations, magnetic fields, and MW pulse step sizes. This sensitivity ratio is an apples-to-apples comparison of the VPDR and DQ Ramsey schemes, both under the same assumption of an approximately known external field such that single- τ measurements are meaningful.

For the full two-dimensional (2D) spectral analysis performed for Figs. 4–7, the VPDR sensitivity is worsened by an additional factor arising from sampling at many nonoptimal free evolution times τ_k . Figure 8(b) shows an example of this factor as a function of the maximum free evolution time, assuming evenly spaced τ_k starting from 0 with $\tau_k < \tau_{\max}$. We again consider the $\langle 1\bar{1}1 \rangle$ orientation with the same parameters as in Fig. 8(a) and a Blackman window function. We add Gaussian noise to $S_{\text{VPDR}}(t_j, \{\tau_k\})$ with $\sigma = 10^{-4}$ and use it to find noisy versions of $f_{\text{VPDR}}(\{\tau_k\})$ and $f_R(\{\tau_k\})$, again reducing the noise in $f_R(\{\tau_k\})$ by $\sqrt{N}$, where N is the number of MW pulse durations used for the VPDR signal. Next we fit the noisy signals $f_{\text{VPDR}}(\{\tau_k\})$ and $f_R(\{\tau_k\})$ to exponentially decaying sinusoids, using a single sinusoid when only $m_I = 0$ is included and using three sinusoids when the full hyperfine triplet is simulated; these fits allow us to extract a (noisy) estimate of the Larmor frequency ω_L . We perform the Monte Carlo calculation 100 times for each $\tau_{\max}$ and estimate the axial magnetic field uncertainty ΔB_p^{fit} from the standard deviation in ω_L . Finally, we take the ratio $\Delta B_p^{\text{fit}} \sqrt{M} / \Delta B_p^{\text{opt}}$, rescaling the noise to account for the number M of free evolution times used in the fit.

Unsurprisingly, the sensitivity cost to fitting many free evolution times (rather than measuring only at τ_{opt}) is the same for VPDR and DQ Ramsey measurements—as expected, since they have the same functional dependence on τ . The cost is higher when hyperfine structure is included, reaching ~ 5 times for the free evolution ranges used in Figs. 4–6. The free-evolution-range cost to sensitivity shown in Fig. 8(b) is representative of what we see for different NV orientations and different magnetic fields (except near the dead zones, where the cost increases). To compare the sensitivity of a full 2D spectrum with that of a DQ Ramsey measurement at $\tau = \tau_{\text{opt}}$, one would need to multiply the factor from Fig. 8(a) by the factor from Fig. 8(b); in this example, the sensitivity would be roughly 30 times worse. Nevertheless, the factor from Fig. 8(a) alone is arguably the fairer comparison, since it makes similar assumptions about the context for VPDR and DQ Ramsey measurements.

Finally, it is worth emphasizing that Eqs. B11–B12 and the results shown in Fig. 8 are specific to the inner product analysis along the t dimension. For example, one might conceivably improve the sensitivity by performing multiple inner products on the same dataset to pick out the double-quantum signals modulated by both Ω and 2Ω , and

extract information from both results. Thus, there is room for future optimization of inversion strategies in both the t dimension and the τ dimension.

2. Sensitivity loss as $\omega_L \rightarrow 0$

As shown in Appendix B 1, the signal sensitivity to changes in the magnetic field depends on $df/d\omega_L \propto s(\tau) = e^{-2\tau/T_2^*} \tau \sin 2\omega_L \tau$, where we have now included an exponential dephasing rate as appropriate for typical ensemble samples. As $\omega_L \rightarrow 0$, $s(\tau) \rightarrow 0$ and the protocol becomes insensitive to changes in the magnetic field.

Practically, the reason for loss of sensitivity as $\omega_L \rightarrow 0$ is that we have not allowed dynamic control over the direction of the MW field. To shift the phase of the sinusoidal oscillation in $f_{\text{VPDR}}(\tau)$ from $\cos 2\omega_L \tau$ to $\sin 2\omega_L \tau$ (which would retain sensitivity at vanishing ω_L) requires one to change the phase of the bright-state superposition $|B\rangle$ targeted by the second microwave pulse relative to the first. While double-quantum Ramsey protocols in a large bias field can spectrally isolate the $|0\rangle \rightarrow |+1\rangle$ and $|0\rangle \rightarrow |-1\rangle$ transitions, allowing dynamic control over the bright-state phase [42], in the absence of a bias field both transitions are targeted by the same MW frequency, and the bright-state phase is determined by the MW polarization. For example, if we changed the direction of the linearly polarized MW field (written in the NV coordinate frame) from $\hat{x}$ during the first pulse to $\cos \theta \hat{x} + \sin \theta \hat{y}$ during the second pulse, then Eq. (2) would be modified to

$$P_0(t, \tau) \approx \cos^4 \frac{\Omega t}{2} + \frac{\cos(\delta\tau + \phi)}{2} \sin^2 \Omega t \cos(\omega_L \tau - \theta) + \sin^4 \frac{\Omega t}{2} \cos^2(\omega_L \tau - \theta). \quad (\text{B13})$$

For $\theta = \pi/4$, we would regain optimal sensitivity at vanishing ω_L . However, given the practical challenge of implementing such a shift in direction while maintaining a constant Rabi magnitude for each orientation, we have restricted our calculations to a constant MW direction.

We can use the limiting behavior of $s(\tau)$ as $\omega_L \rightarrow 0$ to estimate the size of the associated dead zone. While for large $\omega_L \gg 1/T_2^*$ the maximum value of s occurs near $\tau = T_2^*/2$ with a typical magnitude of $T_2^*/2e$, at small $\omega_L \ll 1/T_2^*$ it vanishes linearly with ω_L , i.e., $s(\tau) \approx 2\tau^2 \omega_L e^{-2\tau/T_2^*}$, with a maximum value $s(T_2^*) \approx 2(T_2^*)^2 \omega_L / e^2$. If we require that the magnetometer signal $df/d\omega_L$ stay within a factor of ϵ of its value for large ω_L , we are restricted to operating with $\omega_L > \epsilon(e/4T_2^*)$. For a T_2^* value of 2 μs , we thus require an axial magnetic field $\gtrsim 2\epsilon \mu\text{T}$, which is much less than Earth's magnetic field (25–65 μT). For $\epsilon = 0.25$, the $\hat{z}$ component of a 50 μT magnetic field violates this bound only for angles within $\pm 0.6^\circ$ of the x - y plane. Numerical simulations also support

the conclusion that the angular dead zones are small for magnetic field magnitudes in the range of Earth's field.

APPENDIX C: SIMULATOR APPROXIMATIONS

We consider simulated datasets comprising hundreds to thousands of values of t and τ , corresponding to 10^4 – 10^6 (t, τ) pairs, and compute them for many values of the external magnetic field. To permit the simulations to run in reasonable time on standard computing hardware, we make several approximations that greatly speed up the execution time.

First, we ignore the nonsecular terms of the hyperfine interaction with the ^{14}N nuclear spin. In the absence of a bias field, the transitions driven by the nonsecular terms are strongly suppressed by $\sim A_{\perp}/\Delta$, where $A_{\perp}/(2\pi) = 2.7$ MHz [71] and $\Delta/(2\pi) = 2.87$ GHz is the zero-field splitting. While the resulting small shifts in eigenvalues could impact accurate conversion of $\Delta\omega_i$ to the magnetic field (especially near level anticrossings), such considerations are shared by all magnetometer schemes that detect spin transition frequencies. By ignoring off-axis terms, we can then treat the hyperfine interaction as an effective magnetic field $\hbar A m_I / g\mu_B$ along the NV symmetry axis $\hat{z}$ of each orientation, where $A/(2\pi) = 2.16$ MHz [49] is the parallel component of the hyperfine tensor. In the secular approximation, the ^{14}N nuclear spin evolves negligibly during the pulse sequence since Larmor precession about the external field is suppressed by the quadrupolar splitting, so we approximate m_I as a random constant uniformly distributed between $\{-1, 0, 1\}$. This approximation reduces the dimensionality of the matrix describing the Hamiltonian for each NV orientation from 9×9 to 3×3 .

Next, we approximate the driven evolution in the presence of an off-axis magnetic field by making the rotating wave approximation in the free Hamiltonian eigenbasis. In the laboratory frame, the Hamiltonian for a given NV orientation and nuclear spin projection m_I is

$$\hat{H}_{\text{lab}} = \overbrace{\hbar\Delta\hat{S}_z^2 + (g\mu_B\mathbf{B}_{\text{dc}} + \hbar A m_I \hat{z}) \cdot \hat{\mathbf{S}}}^{\hat{H}_0} + g\mu_B\mathbf{B}_{\text{MW}} \cos(\nu_{\text{MW}}t + \phi) \cdot \hat{\mathbf{S}}, \quad (\text{C1})$$

where the external dc ($\mathbf{B}_{\text{dc}}$) and MW ($\mathbf{B}_{\text{MW}}$) magnetic fields are written in a coordinate system with $\hat{z}||$ NV symmetry axis and we have indicated the free Hamiltonian terms as $\hat{H}_0$. The Schrödinger equation (or master equation) for $\hat{H}_{\text{lab}}$ can be solved by numerical integration in the laboratory frame, but for $\nu_{\text{MW}} \gg \Omega$ it is very inefficient. Instead, we perform the rotating wave approximation as follows: We first diagonalize $\hat{H}_0$, obtaining the matrix of eigenvectors $\hat{V}$ such that $\hat{V}^\dagger \hat{H}_0 \hat{V} = D$, with D diagonal. We then perform a transformation to a rotating frame in the

diagonal basis,

$$\hat{H}_{\text{rot}}^D = \hat{U}^\dagger \hat{V}^\dagger \hat{H}_{\text{lab}} \hat{V} \hat{U} - i\hat{U}^\dagger \frac{d\hat{U}}{dt}, \quad (\text{C2})$$

using a diagonal matrix $\hat{U}$ with entries $e^{-i\nu_{\text{MW}}t}$ at positions corresponding to $m_s = \pm 1$ -like eigenvectors and 1 at the position corresponding to the $m_s = 0$ -like eigenvector of the free Hamiltonian (these assignments are well-defined because we are working in very small magnetic fields $g\mu_B B_{\text{dc}} \ll \hbar\Delta$). We then ignore the remaining time-dependent terms in $\hat{H}_{\text{rot}}^D$. This step ignores counterrotating terms whose effects may become appreciable at the high Rabi frequencies considered, causing Bloch-Siegert shifts of order $\Omega^2/4\Delta \sim \text{MHz}$. We nevertheless drop them because the VPDR protocol is insensitive to detuning. We thus obtain a 3×3 constant-coefficient matrix $\hat{H}_{\text{RWA}}^D$ representing the rotating-wave-approximation Hamiltonian in the free Hamiltonian eigenbasis.

Finally, we include dephasing within a Lindblad master equation description. While quasistatic noise would be a better approximation of experimental conditions, the convolutions its calculation entails are prohibitively slow; Markovian noise is faster to compute and it produces the same exponential Ramsey decay observed in experiments, so we use it to estimate the impacts of dephasing on the protocol. Note, however, that it will not generate the phase shifts observed from quasistatic baths [60], which is why a cosine inner product is sufficient for inversion of numerical simulations, whereas a complex exponential inner product is used for experimental data analysis. For the simulation, we assume Markovian magnetic noise, corresponding to the jump operators $c_0 = | + 1 \rangle \langle + 1 |$, $c_1 = | - 1 \rangle \langle - 1 |$ with equal dephasing rates $\gamma_i = \sqrt{2}/T_2^*$; we represent the jump operators in the free Hamiltonian eigenbasis. Each NV orientation and nuclear spin projection thus obeys

$$\frac{d\rho}{dt} = -\frac{i}{\hbar} [\hat{H}_{\text{RWA}}, \rho] + \sum_i \gamma_i \left(c_i \rho c_i^\dagger - \frac{1}{2} \{ c_i^\dagger c_i, \rho \} \right), \quad (\text{C3})$$

where ρ is the density matrix. Recasting ρ as a vector, we can rewrite Eq. (C3) as a matrix equation $d\rho/dt = \mathcal{L}\rho$, with $\mathcal{L}$ a 9×9 constant-coefficient matrix.

By constructing the simulator to solve first-order linear differential equations, we allow fast simulation of experiments performed over an evenly spaced range of N pulse durations t_j and M interpulse delays τ_k . For each pulse duration $t_j = j\Delta t$, with $j = 0, 1, \dots$, we can find the propagator $U_{\text{Rabi}}(t_j) = (e^{\mathcal{L}\Delta t})^j$ corresponding to evolution under MW driving for duration t_j . Since we wish to calculate all of the $U_{\text{Rabi}}(t_j)$ to simulate all of the pulse durations, we can do it very efficiently by calculating $U_{\text{Rabi}}(t_1) = e^{\mathcal{L}\Delta t}$ once and then recursively multiplying matrices to find $U_{\text{Rabi}}(t_{j+1}) = e^{\mathcal{L}\Delta t} U_{\text{Rabi}}(t_j)$. Similarly,

we can recursively find all of the $U_{\text{Ramsey}}(\tau_k) = (e^{\mathcal{L}_0 \Delta \tau})^k$ where $\mathcal{L}_0$ lacks driving terms. For speed, we do these calculations in the diagonalized basis. Once the propagators are calculated, $P_0(t_j, \tau_k)$ can be determined from the appropriate component of $U_{\text{Rabi}}(t_j)U_{\text{Ramsey}}(\tau_k)U_{\text{Rabi}}(t_j)$.

This calculation is performed for each NV orientation and nuclear spin projection m_I ; averaging over the 12 signals, equally weighted, yields the ensemble outcome P_0^{total} . Finally, having computed P_0^{total} with $\phi = 0$, we repeat the calculation with $\phi = \pi$ and add the results to obtain the SQ-canceled signal S_{VPDR} fed into our data analysis.

APPENDIX D: EXPERIMENTAL FIELD RECONSTRUCTION

In this appendix, we describe how we fit the measured VPDR signals to extract a magnetic field in crystal coordinates that is linearly increasing in coil voltage. From the locations of the inner-product maxima in Fig. 7(c), we obtain $(\omega_{\text{max},i}, \nu_{\text{max},i})$ for one of the $m_I = +1$ features of each orientation i . We estimate the magnitude of the field projection on the i th orientation's symmetry axis $|\beta_i^{\text{exp}}|$ by calculating

$$|\beta_i^{\text{exp}}| = |\hbar(\omega_{\text{max},i} - 2A)/(2g\mu_B)|, \quad (\text{D1})$$

where we subtract the hyperfine splitting to account for our using the $m_i = +1$ feature. This is an approximation valid for small off-axis fields $\beta_{\perp}$ relative to the zero-field splitting, with corrections of order $(g\mu_B)\beta_{\perp}^2/\hbar\Delta$, which even for $\beta_{\perp} = 100 \mu\text{T}$ is $\sim 0.1 \mu\text{T}$. We obtain $|\beta_i^{\text{exp}}(V_n)|$ for the range of coil voltages V_n at which we perform experiments.

Next, we parameterize the external field in the conventional diamond unit cell coordinates ($\hat{x} \parallel \langle 100 \rangle, \hat{y} \parallel \langle 010 \rangle, \hat{z} \parallel \langle 001 \rangle$) as $B_a^{(C)}(V) = b_a + s_a \times V$, where $a \in \{x, y, z\}$, (C) indicates that we are working in crystal coordinates, V is the coil voltage, and $\{b_x, b_y, b_z\}$ and $\{s_x, s_y, s_z\}$ are offset fields and slopes that we seek to determine. The projections of this linear field on each NV axis $\hat{z}_i$ can then be found via $B_i(V) = \hat{z}_i \cdot \vec{B}^{(C)}(V)$. Finally we minimize

$$\chi^2 = \sum_{n,i} (|\beta_i^{\text{exp}}(V_n)| - |B_i(V_n)|)^2 \quad (\text{D2})$$

to obtain the offsets b_a and slopes s_a that best fit a linear voltage dependence for the magnetic field. The best-fit offsets and slopes permit us to calculate both $|B_i(V_n)|$ [as shown in Fig. 7(e)] and $B_a^{(C)}(V)$ [as shown in Fig. 7(f)]. Note that this procedure does not have a unique solution; we arbitrarily pick one choice of the possible permutations of NV axes and magnetic field projection signs by virtue of the initial guesses that we provide to the fit. Nevertheless, even with this degeneracy, the fact that we can find a good fit to all four orientations' signals with three vector field components demonstrates that the VPDR data are mutually consistent.

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