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Geometric phase magnetometry using a solid-state spin

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A key challenge of magnetometry lies in the simultaneous optimization of magnetic field sensitivity and maximum field range. In interferometry-based magnetometry, a quantum two-level system acquires a dynamic phase in response to an applied magnetic field. However, due to the $2\pi$ periodicity of the phase, increasing the coherent interrogation time to improve sensitivity reduces field range. Here we introduce a route towards both large magnetic field range and high sensitivity via measurements of the geometric phase acquired by a quantum two-level system. We experimentally demonstrate geometric-phase magnetometry using the electronic spin associated with the nitrogen vacancy (NV) color center in diamond. Our approach enables unwrapping of the $2\pi$ phase ambiguity, enhancing field range by 400 times. We also find additional sensitivity improvement in the nonadiabatic regime, and study how geometric-phase decoherence depends on adiabaticity. Our results show that the geometric phase can be a versatile tool for quantum sensing applications.
The geometric phase\textsuperscript{1,2} plays a fundamental role in a broad range of physical phenomena\textsuperscript{3–5}. Although it has been observed in many quantum platforms\textsuperscript{6–9} and is known to be robust against certain types of noise\textsuperscript{10,11}, geometric phase applications are somewhat limited, including certain protocols for quantum simulation\textsuperscript{12,13} and computation\textsuperscript{14–17}. However, when applied to quantum sensing, e.g., of magnetic fields, unique aspects of the geometric phase can be exploited to allow realization of both good magnetic field sensitivity and large field range in one measurement protocol. This capability is in contrast to conventional dynamic-phase magnetometry, where there is a trade-off between sensitivity and field range. In dynamic-phase magnetometry using a two-level system (e.g., two spin states), the amplitude of an unknown magnetic field \( B \) can be estimated by determining the relative shift between two energy levels induced by that field (Methods). A commonly used approach is to measure the dynamic phase accumulated in a Ramsey interferometry protocol. An initial resonant \( \pi/2 \) pulse prepares the system in a superposition of the two levels. In the presence of an external static magnetic field \( B \) along the quantization axis, the system evolves under the Hamiltonian \( H = \hbar \gamma B \hat{z}/2 \), where \( \gamma \) denotes the gyromagnetic ratio and \( \hat{z} \) is the z-component of the Pauli spin vector. During the interaction time \( T \) (limited by the spin dephasing time \( T_\text{2*} \)), the Bloch vector \( \mathbf{s}(t) \) depicted on the Bloch sphere precesses around the fixed Larmor vector \( \mathbf{R} = (0, 0, yB) \), and acquires a dynamic phase \( \phi_d = \gamma BT \). The next \( \pi/2 \) pulse maps this phase onto a population difference \( P = \cos \phi_d \), which can be measured to determine \( \phi_d \) and hence the magnetic field \( B \) (Supplementary Note 1).

Such dynamic-phase magnetometry possesses two well-known shortcomings. First, the sinusoidal variation of the population difference with magnetic field leads to a \( 2\pi \) phase ambiguity in interpretation of the measurement signal and hence determination of \( B \). Specifically, since the dynamic phase is linearly proportional to the magnetic field, for any measured signal \( P_{\text{meas}} \) (throughout the text, this value corresponds to \( (\text{AEFL/FL}) \times k \), where \( k \) is a constant that depends on NV readout contrast), there are infinite magnetic field ambiguities: \( B_m = (yT)^{-1} \cos^{-1} P_{\text{meas}} + 2nm \), where \( m = 0, \pm 1, \pm 2 \ldots \pm \infty \). Thus, the range of magnetic field amplitudes that one can determine without modulo \( 2\pi \) phase ambiguity is limited to one cycle of oscillation: \( B_{\text{max}} \propto 1/T \) (Supplementary Note 2, Supplementary Figure 5). Second, there is a trade-off between magnetic field sensitivity and field range, as the interaction time also restricts the shot-noise-limited magnetic field sensitivity: \( \eta \propto 1/T^{1/2} \). Consequently, an improvement in field range via shorter \( T \) comes at the cost of a degradation in sensitivity (Supplementary Note 3). Use of a closed-loop lock-in type measurement\textsuperscript{18}, quantum phase estimation algorithm\textsuperscript{19,20} or non-classical states\textsuperscript{21,22} can alleviate these disadvantages; however, such approaches require either a continuous measurement scheme with limited sensitivity, large resource overhead (additional experimental time) or realization of long-lived entangled or squeezed states.

In the present work, we use the electronic spin associated with a single nitrogen vacancy (NV) color center in diamond to demonstrate key advantages of geometric-phase magnetometry: (i) it resolves the \( 2\pi \) phase ambiguity limiting dynamic-phase magnetometry; and (ii) it decouples magnetic field range and sensitivity, leading to a 400-fold enhancement in field range at constant sensitivity in our experiment. We also show additional improvement of magnetic field sensitivity in the nonadiabatic regime of mixed geometric and dynamic-phase evolution. By employing a power spectral density analysis\textsuperscript{23}, we find that adiabaticity plays an important role in controlling the degree of coupling to environmental noise and hence the spin coherence timescale.

Results

**Geometric-phase magnetometry protocol.** To implement geometric-phase magnetometry, we use a modified version of an experimental protocol ("Berry sequence") previously applied to a superconducting qubit\textsuperscript{8}. In our realization, the NV spin sensor is placed in a superposition state by a \( \pi/2 \) pulse, where the driving frequency of the \( \pi/2 \) pulse is chosen to be resonant with the NV \( m = 0 \leftrightarrow m = +1 \) transition at constant bias field \( B_{\text{bias}} \) (\( \approx 9.6 \) mT in our experiment) aligned with the NV axis. A small signal field \( B \) (\( \sim 100 \) \( \mu \)T in our experiment) is then applied parallel to \( B_{\text{bias}} \), and the NV spin acquires a geometric phase due to off-resonant microwave driving with control parameters cycled along a closed path as illustrated in Fig. 1b (Methods). Under the rotating wave approximation, the effective two-level Hamiltonian is given by:

\[
H = \hbar \left( \Omega \cos(\phi) \sigma_x + \Omega \sin(\phi) \sigma_y + yB \sigma_z \right).
\]  

Here, \( \Omega \) is the NV spin Rabi frequency for the microwave driving field, \( \phi \) is the phase of the driving field, and \( \sigma = (\sigma_x, \sigma_y, \sigma_z) \) is the Pauli spin vector. By sweeping the phase, the Larmor vector \( \mathbf{R}(t) = R^*(\sin \theta \cos \phi, \sin \theta \cos \phi, \cos \theta) \), where \( \cos \theta = yB/(\Omega^2 + (yB)^2)^{1/2} \), rotates around the z-axis. The Bloch vector \( \mathbf{s}(t) \) then undergoes precession around this rotating Larmor vector (for detailed picture of the measurement protocol, see Supplementary Fig. 2). If the rotation is adiabatic (i.e., adiabaticity parameter \( A \equiv \hbar \sin \theta /2\pi \ll 1 \)), then the system acquires a geometric phase proportional to the product of (i) the solid angle \( \Theta = 2\sin(\cos \theta) \) subtended by the Bloch vector trajectory and (ii) the number of complete rotations \( N \) of the Bloch vector around the Larmor vector in the rotating frame defined by the frequency of the initial \( \pi/2 \) pulse. We apply this Bloch vector rotation twice during the interaction time \( T \), with alternating direction separated by a \( \pi \) pulse, which cancels the accumulated dynamic phase and doubles the geometric phase: \( \phi_g = 2N\Theta \) (Supplementary Note 1). A final \( \pi/2 \) pulse allows this geometric phase to be determined from standard fluorescence readout of the NV spin-state population difference:

\[
P_{\text{meas}}(B) = \cos \left[ 4\pi N \left(1 - \frac{yB}{\sqrt{(yB)^2 + \Omega^2}} \right) \right].
\]  

This normalized geometric-phase signal (Supplementary Note 1) exhibits chirped oscillation as a function of magnetic field. There are typically only a small number of field ambiguities that give the same signal \( P_{\text{meas}} \); these can be resolved uniquely by measuring the slope \( dP_{\text{meas}}/dB \) (Supplementary Note 2, Supplementary Fig. 5). From the form of Eq. (2) it is evident that at large \( B \), cosine signal approaches to zero like \( B^{-2} \), and the slope goes to zero. Hence, we define the field range as the largest magnetic field value \( B_{\text{max}} \) that gives the last oscillation minimum in the signal: \( B_{\text{max}} \propto \Omega N^{1/2} \). Importantly, the field range of geometric-phase magnetometry has no dependence on the interaction time \( T \). If the magnetic field is below \( B_{\text{max}} \), then one can make a geometric-phase magnetometry measurement with optimal sensitivity \( \eta \propto \Omega^{-1/2} \) (Supplementary Note 3).

**Comparison between dynamic- and geometric-phase magnetometry.** We implemented both dynamic- and geometric-phase magnetometry using the optically addressable electronic spin of a single NV color center in diamond (Fig. 2a) (Supplementary Figs. 1-3). NV-diamond magnetometers provide high spatial...
resolution under ambient conditions \(24-26\) and have therefore found wide-ranging applications, including in condensed matter physics \(27,28\), the life sciences \(29,30\), and geoscience \(31\). At an applied bias magnetic field of 9.6 mT, the degeneracy of the NV spin \(m_z = \pm 1\) is lifted. The two-level system used in this work consists of the ground state magnetic sublevels \(m_z = 0\) and \(m_z = \pm 1\), which can be coherently addressed by applied microwave fields. The hyperfine interaction between the NV electronic spin and the host \(^{14}\)N nuclear spin further splits the levels into three states, each separated by 2.16 MHz. Upon green laser illumination, the NV center exhibits spin-state-dependent fluorescence and optical pumping into \(m_z = 0\) after a few microseconds. Thus, one can prepare the spin states and determine the population by measuring the relative fluorescence (see Methods for more details).

First, we performed dynamic-phase magnetometry using a Ramsey sequence to illustrate the \(2\pi\) phase ambiguity and show how the dependence on interaction time gives rise to a trade-off between field range and magnetic field sensitivity. We recorded the NV fluorescence signal as a function of the interaction time \(T\) between the two microwave \(\pi/2\) pulses (Fig. 1a). Signal contributions from the three hyperfine transitions of the NV spin result in the observed beating behavior seen in Fig. 2b. We fixed the interaction time at \(T = 0.2, 0.5, 1.0\) \(\mu\)s, varied the external magnetic field for each value of \(T\), and observed a periodic fluorescence signal with a \(2\pi\) phase ambiguity (Fig. 2c). The oscillation period decreased as the interaction time was increased, indicating a reduction in the magnetic field range (i.e., smaller \(B_{\text{max}}\)). In contrast, the magnetic field sensitivity, which depends on the maximum slope of the signal, improved as the interaction time increased. For each value of \(T\), we fit the fluorescence signal to a sinusoid dependent on the applied magnetic field and extracted the oscillation period and slope, which we used to determine the experimental sensitivity and field range. From this procedure, we obtained \(\eta \propto T^{-0.49(6)}\) and \(B_{\text{max}} \propto T^{-0.96(2)}\), consistent with expectations for dynamic-phase magnetometry and illustrative of the trade-off inherent in optimizing both \(\eta\) and \(B_{\text{max}}\) as a function of interaction time (Supplementary Fig. 7).
Next, we used a Berry sequence to demonstrate two key advantages of geometric-phase magnetometry: i.e., there is neither a $2\pi$ phase ambiguity nor a sensitivity/field-range trade-off with respect to interaction time. For fixed adiabatic control parameters of $\Omega/2\pi = 5$ MHz and $N = 3$, the observed geometric-phase magnetometry signal $P_{\text{meas}}$ has no dependence on interaction time $T$ (Fig. 2d). Varying the external magnetic field with fixed interaction times $T = 4.0, 6.0, 8.0 \mu$s, $P_{\text{meas}}$ exhibits identical chirped oscillations for all $T$ values (Fig. 2e), as expected from Eq. (2). From the $P_{\text{meas}}$ data we extract $dP_{\text{meas}}/dB$, which allows us to determine the magnetic field uniquely for values within the oscillatory range (Supplementary Note 2), and also to quantify $B_{\text{max}}$ from the last minimum point of the chirped oscillation (Fig. 2e). Additional measurements of the dependence of $P_{\text{meas}}$ on the adiabatic control parameters $\Omega$, $N$, and $T$ (Supplementary Figs. 4, 6) yield the scaling of sensitivity and field range: $\eta \propto \Omega^{1.2(5)}N^{-0.92(1)}T^{0.46(1)}$ and $B_{\text{max}} \propto \Omega^{0.9(1)}N^{0.52(5)}T^{0.02(1)}$, which is consistent with expectations and shows that geometric-phase magnetometry allows $\eta$ and $B_{\text{max}}$ to be independently optimized as a function of interaction time (Supplementary Fig. 7).

In Fig. 3 we compare the measured sensitivity and field range for geometric-phase and dynamic-phase magnetometry. For each point displayed, the sensitivity is measured directly at small $B$ (0.01 ~ 0.1 mT), whereas the field range is calculated from the measured values of $N$ and $\Omega$ (for geometric-phase magnetometry) and $T$ (for dynamic-phase magnetometry, with $T$ limited by the dephasing time $T_2^*$), following the scaling laws given above. Since geometric-phase magnetometry has three independent control parameters ($T$, $N$, and $\Omega$), $B_{\text{max}}$ can be increased without changing sensitivity by increasing $N$ and $\Omega$ while keeping the ratio $N/\Omega$ fixed. Such “smart control” allows a tenfold improvement in geometric-phase sensitivity (compared to dynamic-phase measurements) for $B_{\text{max}} \sim 1$ mT, and a 400-fold enhancement...
interaction time at sensitivities provided by dynamic-phase magnetometry, we unavoidably coupled as deviation of the results.

Geometric-phase magnetometry in nonadiabatic regime.

Finally, we explored geometric-phase magnetometry outside the adiabaticity controls the coupling between the NV spin and environmental noise during geometric manipulation, thereby determining the geometric-phase coherence time. Furthermore, we showed that operation in the nonadiabatic regime, where there is mixed geometric- and dynamic-phase evolution, allows magnetic field sensitivity to be better than that of dynamic-phase magnetometry. We expect that geometric-phase AC field sensing will provide similar advantages to dynamic-phase magnetometry, although the experimental protocol (Berry sequence) will need to be adjusted to allow only accumulation of geometric phase due to the AC field. The generality of our geometric-phase technique should make it broadly applicable to precision measurements in many quantum systems, such as trapped ions, ultracold atoms, and other solid-state spins.

Geometric-phase coherence time.

Figure 4b shows examples of the measured decay of the geometric-phase signal ($P_{\text{max}}$) as a function of interaction time $T$ and adiabaticity parameter $A$. From such data we extract the geometric-phase coherence time $T_2g$ by fitting $P_{\text{max}} \sim \exp(-(T/T_{2g})^2)$. We observe four regimes of decoherence behavior (Fig. 4c), which can be understood from Eq. (3) and its schematic spectral representation in Fig. 4d. For $A < 0.1$ (adiabatic regime), dynamic-phase evolution (i.e., Hahn-echo-like behavior) dominates the decoherence function $\chi(T)$ and thus $T_{2g} \sim T_s = 500 \mu s$. For $0.1 \leq A < 1.0$ (intermediate regime), the coherence time is inversely proportional to the adiabaticity parameter ($T_{2g} \sim 1/A$) as geometric-phase evolution (with Ramsey-like dephasing) becomes increasingly significant. For $A \approx 1.0$ (nonadiabatic regime), geometric-phase evolution dominates $\chi(T)$ at long times and thus $T_{2g} \sim T_s^* = 50 \mu s$. For $A > 1.0$ (strongly nonadiabatic limit), the driven rotation of the Larmor vector is expected to average out during a Berry sequence (Fig. 4b) and only the $z$-component of the Larmor vector remains. Thus, the Berry sequence converges to a Hahn-echo-like sequence and the coherence time is expected to increase to $T_s$ for very large $A$.

Discussion.

In summary, we demonstrated an approach to NV-diamond magnetometry using geometric-phase measurements, which avoids the trade-off between magnetic field sensitivity and maximum field range that limits traditional dynamic-phase magnetometry. For an example experiment with a single NV, we realize a 400-fold enhancement in static (DC) magnetic field range at constant sensitivity. We also explored geometric-phase magnetometry as a function of adiabaticity, with good agreement between measurements and model simulations. We find that adiabaticity controls the coupling between the NV spin and environmental noise during geometric manipulation, thereby determining the geometric-phase coherence time. Furthermore, we showed that operation in the nonadiabatic regime, where there is mixed geometric- and dynamic-phase evolution, allows magnetic field sensitivity to be better than that of dynamic-phase magnetometry. We expect that geometric-phase AC field sensing will provide similar advantages to dynamic-phase magnetometry, although the experimental protocol (Berry sequence) will need to be adjusted to allow only accumulation of geometric phase due to the AC field. The generality of our geometric-phase technique should make it broadly applicable to precision measurements in many quantum systems, such as trapped ions, ultracold atoms, and other solid-state spins.
Methods

NV diamond sample. The diamond chip used in this experiment is an electronic-grade single-crystal cut along the [110] direction into a volume of 4 x 4 x 0.5 mm³ (Element 6 Corporation). A high-purity chemical vapor deposition layer with 99.99% 12C near the surface contains preferentially oriented NV centers. The estimated N and NV densities are 1 x 10^{15} and 3 x 10^{13} cm⁻³, respectively. The geometric phase coherence time and sensitivity in nonadiabatic regime. a Measured geometric-phase magnetic field sensitivity-squared (red squares) plotted against adiabaticity parameter A for a fixed interaction time of T = T₂/2 at which the dynamic-phase Ramsey sequence gives optimal sensitivity (dashed blue line). Dashed red line shows geometric-phase sensitivity lower limit calculated by a numerical simulation assuming maximum signal contrast. The simulation does not include the contrast reduction due to hyperfine modulation. b Measured Berry sequence signal as a function of interaction time T for various adiabaticity parameter values. Color dots are data; solid color lines are exponential fits to data – exp(T/T₂)². Blue and green dashed lines indicate T₂⁺ and T₂⁻ decay of the dynamic-phase signal measured with a Ramsey and Hahn-echo sequence, respectively. c Measured geometric-phase coherence time T₂ for a function of adiabaticity parameter A. Three regimes are observed: (i) For A < 0.1, T₂ ≈ T – T₂⁻, (ii) For 0.1 < A < 1.0, T₂⁻ = T₂⁺ ~ 50 µs, and (iii) For A > 1.0, T₂⁻ ≈ T₂⁺ ~ 50 µs. d Qualitative representation of contributions to the decoherence function (Eq. (3)) in the frequency domain: environmental noise spectral density function S(ω) (black line); dynamic-phase (spin-echo) filter function F₂(ωT)/ω² (dashed green line); and geometric-phase (Berry sequence) filter function A²F₂(ωT)/ω² (dashed blue line) in the limit A → 1. Error bars represent one standard deviation of the results when the spin is in the m_s = 0 state is also measured as a reference. The temperature of the confocal scanning laser microscope is monitored by a 10k thermistor (Thorlabs) and stabilized to within 0.05 °C using a 15 W heater controlled with a PID algorithm.

Hamiltonian parameter control system. The Rabi frequency (Ω) and phase (ϕ) of the microwave drive field, as well as the applied magnetic field to be sensed (B), are key variables of this work. It is thus crucial to calibrate the microwave driving system and magnetic field control system beforehand. Microwave pulses for NV geometric phase magnetometry are generated by mixing a high frequency (~3 GHz) local oscillator signal and a low frequency (~50 MHz) arbitrary waveform signal using an IQ mixer (Supplementary Fig. 1). The Rabi frequency and microwave phase are controlled by the output voltage of an arbitrary waveform generator (Tektronix AWG5014C) (Supplementary Fig. 2). A microwave waveguide (10 µm gap, 1 µm height) fabricated on a glass coverslip by photolithography. An external magnetic field for magnetometry demonstration is created by sending an electric current through a copper electromagnetic coil (4 mm diameter, 0.2 mm thick, n = 40 turns, R = 0.25 Ω) placed h = 0.5 mm above the diamond surface. The electric current is provided by a high-stability DC voltage controller (Agilent E3640A). To enable fine scan of the electric current with limited voltage resolution, another resistor with 150 Ω is added in series. Thus, a DC power supply voltage of 3 V approximately corresponds to I = 0.02 A, which creates an external field of B = μ₀NIh/4πn = 16 G. One can determine the change of the external magnetic field as a function of DC power supply voltage ΔB(V) by measuring the shift of the resonance peak Δf in the NV electron spin resonance spectrum using Δf = γΔB. The result is ΔB/V = 0.50 ± 0.01 G V⁻¹ (Supplementary Fig. 3). Joule heating produced by the coil is P = F R - 10⁻⁴ W. The mass and heat capacity of the coil are about 0.15 g and 0.06 kJ °C⁻¹, respectively. Thus, the temperature rise is at most 2 mK s⁻¹. Since the temperature coefficient of the fractional resistivity change for copper is 0.00386 K⁻¹, the change of resistance due to Joule heating is negligible.
Numerical methods for geometric phase simulation. All simulations of NV spin evolution in this work are carried out by computing the time-ordered time evolution operator at each time step.

$$U(t_i, t_f) = \hat{T} \left( \exp \left( -i \int_{t_i}^{t_f} H(t) \, dt \right) \right) = \prod_{n=1}^{N} \exp \left( -i\hbar H_{\text{t}}(t_\text{n}) \right).$$

where $$t_\text{i}$$ and $$t_\text{f}$$ are the initial and final time, respectively, $$\hat{T}$$ is the time-ordering operator, $$\Delta t$$ is the time step size of the simulation, $$N = (t_\text{f} - t_\text{i}) / \Delta t$$ is the number of time step, and $$H(t)$$ is the time-dependent Hamiltonian (Eq. (1)). In the simulation, we used $$\Delta t$$=1 ns step size which is sufficiently small in the rotating frame. The algorithm is implemented with MATLAB.

Data and code availability. The data and numerical simulation code that support the findings of this study are available from the corresponding author upon reasonable request.

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