Composite-pulse enhanced room-temperature diamond magnetometry

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ABSTRACT
The sensitivity of practical solid quantum sensing can be boosted up by increasing the number of probes. However, the effects of spin dephasing caused by inhomogeneous broadening and imperfect quantum control can reduce the fidelity of quantum control and the sensitivity of quantum sensing with the dense ensemble of probes, such as nitrogen-vacancy (NV) centers in diamond. Here, we present a robust and effective composite-pulse for high fidelity operation against inhomogeneous broadening and control errors via optimized modulation of the control field. Such a composite-pulse was verified on NV center to keep high fidelity quantum control up to a spectrum detuning as large as 110\% of Rabi frequency. The sensitivity of the magnetometer with NV center ensemble was experimentally improved by a factor of 4, comparing to dynamical decoupling with a normal rectangular pulse. Our work marks an important step towards high trustworthy ultra-sensitive quantum sensing with imperfect quantum control in practical applications. The used principle is universal and not restricted to NV center ensemble magnetometer.

1. Introduction
Solid-state electronic spin defects, including nitrogen vacancy (NV) [1], silicon-vacancy [2] and germanium-vacancy [3] centers in diamond, have garnered increasing relevance for quantum science and modern physics metrology. Especially, NV centers have been systematically studied and employed in interdisciplinary applications facilitated by long electron spin coherence time [4–6] at room-temperature. Although, a single NV center can be used for high-spatial-resolution field imaging [7, 8], enhancing sensitivity are feasibly achieved by using an ensemble of \( N \) NV centers, where the shot-noise limited magnetic field sensitivity can be improved by a factor \( \sqrt{N} \) [5, 9, 10]. There have been many applications utilizing dense NV center ensembles for high-sensitivity magnetic-field sensing [9] and wide-field magnetic imaging [11], such as measurements of single-neuron action potential [12], current flow in graphene [13] and chemical shift spectra from small molecules [14].

However, for a quantum magnetometry based on NV center ensembles as shown in Figure 1, the detection sensitivity is usually limited by quantum dephasing process of electron spin [15]. The typical electron spin dephasing time is less than 1 \( \mu \text{s} \), which is primarily caused by the spectrum detuning (\( \delta \)) due to inherent or extrinsic inhomogeneous broadening [15–20]. Randomly localized \( ^{13} \text{C} \) nuclei spins, nitrogen impurities (P1 centers) [6, 21], other unknown spins, strain gradients, magnetic-field gradients, and temperature fluctuations contribute to NV center dephasing process. Moreover, for the near-surface NV center in bulk diamond or NV center in nano-diamond, the electric field noise from surface charge fluctuations can be another part of spin decoherence and induce a significant inhomogeneous broadening [22]. Beside those intrinsic dephasing effects, detunings, and inhomogeneous in the microwave frequency and amplitude also destroy the coherence of system. With increasing the number of imperfect quantum controls, more rapid control errors accumulate and even generate spurious signals in quantum sensing process [23–25].

One particularly successful approach to reducing the above imperfections developed from nuclear magnetic resonance experiment is the use of composite pulses [26, 27], in which a single rotation about some axis in the x-y plane is replaced by a series of rotations. In such a way,
their errors can be canceled over the sequence. More recently, this method has been used for quantum computation to improve the operation fidelity in solid system [28–30]. In this work, we showed an effective dynamical composite-pulse operation capable of achieving extensive robust and high sensitive quantum magnetometry based on NV center ensembles in diamond. By making use of gradient ascent algorithm [31, 32], we can parameterize the phase and duration of control pulse and optimize them with employing experimental testing to meet high fidelity and robustness requirements. On NV center ensemble, the optimized composite-pulse sequence can keep high fidelity operation for a normalized detuning as large as 110 % of Rabi frequency and improve the sensitivity by a factor of 4 comparing to dynamical decoupling (DD) with normal rectangular pulse. The physical mechanisms behind of the enhancement for the optimized composite-pulse was also presented with theoretical and experimental study by investigating the quantum dynamically temporal evolution process of the sensing protocol. Such a simple and robust method for the NV centers ensemble quantum magnetometry outlines a path towards effective and trustworthy quantum sensing.

2. Materials and methods

NV ensemble magnetometry experiments were performed on a chemical vapor deposition (CVD) grown diamond with [N] ⊗2 ppm and [NV ] [30.0063 ppm (described in Appendix A). The sample was grown on a type Ib commercial high-pressure high temperature (HPHT) (100)-oriented single crystal diamond with approximate dimensions 3×3×0.5 mm³ from Element-6. Before we used the diamond substrates, they were cleaned in a mixture of sulfuric and nitric acid (1:2) for 1 h at 200°C and rinsed with de-ionized water, acetone, and isopropanol. Before growth, the diamond substrates were pre-treated using an plasma in order to prepare their surfaces for single crystal diamond growth. The microwave plasma CVD system (MPCVD, Seki Technnotron Corp. AX-5250S) was used to grow the samples with the microwave power of 4.5 ~ 5.0 kW at a pressure of 140 ~ 160 Torr. The growth temperature was about 850 ~ 1000 °C. Hydrogen (5 N), 6% CH₄ (5 N), and no intentional addition of nitrogen (N₂) were used. After growth, the sample was separated from the HPHT diamond substrate by laser cutting. Both sides of the growth plates were polished by a mechanical polisher. We studied naturally occurring NV centers that were located 5 µm below the surface. We used a room-temperature home-built confocal microscopy with a dry objective lens (N.A. = 0.9) to address NV centers. In the experiment, we made use of power splitters and phase shifters to control the relative phases between different microwave (MW) channels. And we calibrated the phase shift with a vector network analyzer by beating each of the channels with a proper attenuation.

The NV center consists of a substitutional nitrogen atom adjacent to a carbon vacancy in diamond crystal lattice, as shown in Figure 1(a). The ground state of NV center is an electron spin-triplet state \( \frac{1}{2} \), and the zero-field splitting between \( m_s = 0 \) and degenerate \( m_s = ±1 \) sub-levels is \( D \otimes 2.87 \) GHz. The electron spin of NV center can be optically initialized into the ground state \( |m_s = 0\rangle \) through the inter-system crossing (ISC). The spin-dependent photon luminescence (PL) enables the implementation of optically detected magnetic resonance (ODMR) technique to detect the spin state. In the experiment, we applied an external magnetic field near the excited state level anti-crossing, i.e. \( B_0 \otimes 250 \) G along the NV symmetry axis to split the energy levels and polarize intrinsic \( ^{14}N \) nuclear spin to enhance the signal contrast [6, 9, 33], as shown in Figure 2(a). Here, the spin-lattice relaxation time of NV center \( T_1 \) was larger than 1 ms, indicating no obvious frequency crosstalk between NV centers and P1 centers as shown in Figure 2(b). Under this large magnetic field, the inhomogeneous broadening effect from strain or electric field can be suppressed [34, 35].

When we apply a single MW to address the transition \( |m_s = 0\rangle \leftrightarrow |m_s = 1\rangle \), the Hamiltonian of NV center can be expressed as \( \mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{int}} + \mathcal{H}_I \), where \( \mathcal{H}_0 = \sum_i \mathbf{A}_i \sigma^z_i \) is the dark spin bath Hamiltonian. Due to large energy mismatching, the term of inhomogeneous broadening can be expressed as \( \mathcal{H}_{\text{int}} = \sigma^z \sum_i \mathbf{A}_i \sigma^z_i \), where \( \sigma^z \) is the spin operator of the ith spin and \( \mathbf{A}_i \) is the strength of the hyperfine interaction between the probe qubit and ith spin. The hyperfine interaction with dark spin results in a random local magnetic field \( \mathbf{B}_l = \sum_i \mathbf{A}_i \sigma^z_i \) with a typical linewidth \( \Gamma_l \) = 2 MHz for NV ensemble as shown in Figure 1(b).
Figure 2. (a) ODMR spectrum of NV center ensemble under $B \approx 500$ G magnetic field. (b) The longitudinal depolarization process of NV center ensemble. (c) Ramsey fringe for total inhomogeneous broadening. (d) Spin-echo sequence for pure dephasing process. The decay law [5] for coherence of NV centers can be characterized by $\exp(-\alpha t) = \exp(-\nu t)^5$ with $\nu \in [3, 6]$ and we chose $\nu = 3$ for our diamond sample.

Figure 2(c). The transverse relaxation time of NV center ($T_2 = 94 \pm 2$ $\mu$s) was measured by spin-echo sequence, as shown in Figure 2(d). Because the typical coherent control is much faster than dynamical fluctuations, we take $\delta$ as a random time independent variable [5, 32]. Furthermore, we assume that NV ensemble dephasing mechanisms are independent and inhomogeneous broadening effect can be described by a Gaussian linetype $P(\delta)$ [32]. Since the fluctuation of MW’s power is always smaller than inhomogeneous broadening in experiment [30, 32], we focus on the fidelity optimization of coherent operation with large detuning. More explicitly, we take the target function [32]

$$F(\xi, U) = \int d\psi \langle \psi | U \xi | \psi \rangle \langle \psi | U | \psi \rangle,$$

where $\xi$ and $U$ are the practical and ideal operations, $|\psi\rangle$ is an arbitrary pure state on Bloch sphere. The average fidelity ($F(\xi, U)$) is a measure of how well $\xi$ approximates a quantum gate, $U$. Note that $F(\xi, U) = 1$ if and only if $\xi$ implements $U$ perfectly, while lower values indicate that $\xi$ is a noisy implementation of $U$. The integral is over the uniform measure $d\psi$ on state space, normalized by $\int d\psi = 1$. The practical quantum operation can be written as $\xi(\psi \langle \psi |) = \int P(\delta)G(\epsilon)U_{\alpha \alpha}(\delta, \epsilon)|\psi\rangle \langle \psi | U_{\alpha \alpha} d\delta d\epsilon$, where $\epsilon$ is the control amplitude fluctuation, and $G(\epsilon)$ is the fluctuation of MW’s power. We assume $\epsilon$ satisfies a Lorentzian distribution [32]. For single qubit operation, the average gate fidelity [32, 36] can be simplified to $F(\xi, U) = \frac{1}{2} + \frac{1}{12} \sum_{\sigma \neq \alpha = x, y, z} \text{Tr} \left( U \sigma_\alpha U^\dagger \xi(\sigma_\alpha) \right)$.

Usually, $\frac{\pi}{2}$ and $\pi$ pulses are employed to form quantum sensing protocol [5, 27, 37]. Furthermore, the $\pi$ pulse, which acts as the core of DD method, directly affects the final magnetometry sensitivity of NV center [24]. Based on the dynamical correct core of quantum control [28, 32], we designed an effective dynamically composite-$\pi$ -pulses with two 5-piece pulses, where $R(\pi) = R^2(\frac{\pi}{2}) = [R_{\alpha \beta}(\frac{\pi}{2})R_{\gamma \delta}(\pi)R_{\alpha \beta}(\frac{\pi}{2})R_{\gamma \delta}(\pi)]^2$ and $\phi$, for $i=1,2,3$, is the initial phase of MW. With gradient ascent algorithm, the optimized relative phases were $\phi_2 - \phi_1 = 97.08^\circ$ and $\phi_3 - \phi_1 = -47.88^\circ$, as shown in Figure 3(a). A typical result with such control composite-pulses is shown in Figure 3(c). For comparison, we also performed a simulation with a rectangular $\pi$ -pulse of the same Rabi frequency as shown in Figure 3(b). The composite pulses enhanced the coherent operation fidelity over a large range of detuning and control amplitudes. Then we applied the pulse sequence on a single NV center. As shown in Figure 3(d), we find good agreement with experimental results as theoretical result. The little divergence was caused by MW’s power or relative phase drifts. The fidelity can keep at 0.9 with the detuning as much as 110% of the Rabi frequency, which is quite better than the rectangular pulse results. Such a control composite-pulse forms the core of quantum sensing protocol of NV center.

3. Results and discussion

Based on the high fidelity and robustness of dynamical composite-pulse control on single NV center, we
employed it with the ensemble of inhomogeneously broadened NV centers for quantum magnetometry. We performed alternating current (AC) magnetic field sensing protocol by employing DD method [5, 9, 27, 37]. The quantum lock-in sequence is shown in Figure 4(a). To characterize the behavior of such a sensor, we also applied a square wave magnetic field in phase with the spin-echo sensing sequence. The typical spin-echo magnetometry results with rectangular pulse were measured by changing the amplitude of AC magnetic field and sensing time as shown in Figure 4(b) without detuning. Under a fixed unknown AC magnetic field, the accumulated relative phase increases linearly with the sensing time and the signal contrast decreases because of inhomogeneous broadening as shown in Figure 4(c). The best sensitivity [5, 38, 39] using the quantum lock-in protocol was at \( v = 87 \), where the variable \( v \) is the index of the decay law for NV center coherence and we chose \( v = 3 \) for our NV ensemble. For a fixed sensing time, the accumulated phase has a linear relationship with the amplitude of AC magnetic field as shown in Figure 4(d). In general, the sensitivity [39] of a magnetic field measurement is given by \( \eta \approx \frac{\sigma}{dS/dB_{\text{un}}} \sqrt{\tau} \), where the standard deviation (\( \sigma \)) of the sensing signal is compared to the response of the system \( dS \) in a changing magnetic field \( dB_{\text{un}} \). So by fitting experimental data, we can get \( dS/dB_{\text{un}} = k \) and \( \sigma \), which is the photon fluctuation as shown in Figure 4(d) with error bar.

Then we scanned the detuning of NV center ensemble over a large range. And for comparison, the final results are shown in Figure 4(e). We can see that the sensitivity with composite-pulses keeps constant up to the detuning of 110% of the Rabi frequency, agreeing with theoretical prediction. However, for the rectangular pulses, it degenerates quickly when the detuning increases over 50% of the Rabi frequency and the uncertainty of sensitivity increases sharply. As detuning further increases for rectangular case, the performance of NV center ensemble is reduced and the fluctuation of sensitivity becomes large. With optimal composite-pulse control, we obtained a stable magnetometric sensitivity times root sensing volume of \( 8 \text{nTHz m}^{-3/2} \), which is mainly limited by the density and coherence time of NV centers in the present diamond sample. When the detuning increases to 110% of the resonant Rabi frequency, we experimentally improved the sensitivity by a factor of 4, comparing to the control with normal rectangular pulse, as shown in Figure 4(f).

To figure out the physical mechanisms behind of the enhancement for the optimized composite-pulse, we investigated the quantum dynamically temporal evolution process of sensing protocol both in theory and experiment as shown in Figure 5(a) and (b). The Hamiltonian of interaction between NV center and nuclear spin can be written as [40] as:

\[
H = D\hat{S}_z - y B_z \hat{S}_x - \sum_n \gamma_n B_z \cdot g_n \langle \hat{S}_x \rangle \hat{I}_n + \sum_n \hat{S}_z A_n \cdot \hat{I}_n + \sum_{n,m} C_{nm} \langle \hat{S}_z \rangle \hat{I}_m.
\]

Figure 3. (a) General rectangular and composite sequence for \( \pi \)-pulse. (b, c) Simulated quantum control fidelity of rectangular (b) and composite (c) dynamical pulses for a range of detuning and control amplitude, scaled by Rabi frequency. Navy lines are contour lines at a fidelity of 0.9. (d) Measured (dots) and simulated (solid lines) fidelity of \( \pi \) pulse. Red (gray) dots correspond to composite (rectangular) pulses.
where $D$ is the zero-field splitting, $\hat{S}$ is the spin-1 operator of the NV center electrons, $\hat{I}_n$ is the spin-$\frac{1}{2}$ operator of the $^{13}$C nucleus, $\gamma_e$ and $\gamma_N$ are the gyromagnetic ratios of the electron spin and the nuclear spin, $g_e$ is the effective $g$ tensor, $B_z$ is the external magnetic field, and $A_n$ is the hyperfine coupling tensor (a $3 \times 3$ matrix) for the $n$th nuclear spin and electron spin, and $C_{nm}$ is the hyperfine coupling tensor (a $3 \times 3$ matrix) for the $n$th and $m$th nuclear spins. In the above Hamiltonian, the second and third term are Zeeman effects for electron and nuclei, the fourth term is the hyperfine interaction between the electron and nuclear spin bath, the fifth term is an effective crystal-field splitting felt by the
nuclear spin bath, and the last term is the dipolar interaction among spin bath. The relatively large zero-field splitting $D$ does not allow the electron spin to flip and thus we can make the so-called secular approximation, removing all terms which allow direct electronic spin flips. Nonsecular terms have been included up to second order in perturbation theory, leading to the $S_z$ dependence of other terms in the Hamiltonian. The sensing signal can be expressed as [5]

$$s = \text{Tr} \left[ P e^{-iH_1t/H} R(\pi) e^{-iH_1t/H} \rho e^{iH_1t/H} R(\pi) e^{iH_1t/H} \right],$$

where the $\rho = \rho_{NV} \otimes \rho_i$ is the probe state, $P$ is projection measurement operator and

$$H_i = (\delta + \gamma B_m) + \sum_n \Omega_n^{(m,n)} \hat{I}_n + \sum_{n \neq m} \tilde{C}_{m,n} \hat{S}_n \cdot \hat{I}_m,$$

$$H_f = (\delta - \gamma B_m) + \sum_n \Omega_n^{(f,n)} \hat{I}_n + \sum_{n \neq m} \tilde{C}_{m,n} \hat{S}_n \cdot \hat{I}_m$$

are the projected Hamiltonian [40] under an unknown AC magnetic field, where $\Omega_n$ denotes the electron spin state, the effective Larmor vector for nuclear spin n, and $C_{m,n}$ is the effective coupling between nuclear spin n and m. So we have

$$s = C + E \text{Re} \left[ \text{Tr} \left( U_f U_i \rho_{NV} U_i^\dagger \right) \right] \cos(2\gamma B_m t + \phi_i) + M$$

where the modulation caused by detuning can be written as

$$M = D \text{Re} \left[ \text{Tr} \left( U_f^\dagger \rho_{NV} U_i \right) \right] \cos(2\gamma B_m t + \phi_i) + \text{Re} \left[ \text{Tr} (U_f \rho_{NV} U_i) \right]$$

and $U_f (U_i)$ is the dark spin Feynman propagator conditioned on the NV electron spin state and can be specialized by cluster-correlation expansion theory [41]. A, B, C, D, and E are constant coefficients. Hence, the amplitude of modulation caused by detuning effect decays with the time scale of dephasing time ($T_2^*$). However, the sensing signal decays with the time scale of $T_2^*$ due to DD method. Due to immunity to detuning, the modulation term of composite-pulse sequence is quite small and can be canceled out by high fidelity of $\pi$-pulse operation as shown in Figure 5(b). But for the rectangular pulse, the fidelity of $\pi$-pulse operation degenerates seriously with detuning when $\delta / \Omega_{\omega} > 50\%$ as shown in Figure 3(d). The detuning effect on the modulation term is significant. It decays fast with the time scale of $T_2^*$ as shown in Figure 5(a). So the sensitivity of rectangular pulse sequence degenerates quickly in the signal contrast for detuning as shown in Figure 4(e).

We also evaluated the multipulse sequence for magnetic field sensing as shown in Figure 6(a). The sensitivity can be further improved by a factor of 2 when the number of $\pi$-pulses is greater than 8. And the relative magnetometry sensitivity enhancement between composite and rectangular sequence increases sharply with the number of $\pi$-pulses and the value of detuning, as shown in Figure 6(b). From the theoretical simulation, the performance of the composite-pulse cooperating with multipulse is much better than that of rectangular pulse at large detuning, corresponding to large spectrum broadening in practical applications. In the experiment, the coherence time of NV center ensemble was $T_2^* \Theta 94\,\mu s$, which was mainly limited by P1 center in diamond. However, with advanced quantum control of P1 center [15], the coherence time of NV center ensemble can be extended to $T_2^* \Theta 12\,\text{ms}$ for samples with P1 center concentration $\sim 1$ ppm [42]. Furthermore, the generating efficiency of NV center ensemble was less than 1% for the diamond sample. The density of NV center can be enhanced more than 150 times [15] by electron irradiation treatment. By applying composite-pulse cooperating with multipulse sequence on the improved NV ensemble, the magnetic

![Figure 6](image-url)

**Figure 6.** (a) Improved magnetometric sensitivity under multipulse sequence for dynamical composite-pulse. The white lines are contour lines at sensitivity $4 \times 10^{-3}$ Hz $\mu m^{-1}$. (b) The relative sensitivity enhancement of composite-pulse sequence compared to rectangular pulse for multipulse sequences.
sensitivity of NV center ensemble can be enhanced towards sub-1 pTHz $^{12}$m/s$^{12}$ even with large inhomogeneous broadening.

4. Conclusions

In this work, we showed that the errors of a wide range inhomogeneous broadening or detuning in quantum sensing can be suppressed by a composite pulse. The absolute sensitivity achieved in our experiment was limited by the density of the NV centers, dephasing time, and the readout contrast, which can be further improved by optimizing fabrication and material parameters. By combining with large area ring-shaped resonators [43], the magnetometry based on NV center ensemble allows for detecting proton spins in a microscopically resolvable volume, bringing new fields of application into reach: for example, noninvasive in vivo sensing of biomagnetic fields and microfluidic chemical analysis, as well as cellular and neuronal action imaging [12], where characteristic dimensions are at the micron scale.

Disclosure statement

No potential conflict of interest was reported by the author(s).

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Appendix A: Experimental condition

For magnetometry applications, we investigate the behavior of the magnetic field sensitivity as a function of the optical pumping power as shown in Figure A1(a). Generally, the ODMR contrast and linewidth are function of the microwave and optical pumping power. And both of them have direct effect on the performance of NV centers magnetometry. As shown in Figure A1(a), the sensitivity of pulsed ODMR is enhanced by increasing the power of 532 nm laser. So in the experiment, we set the power of 532 nm laser of 12 mW and initialize NV center into ground with $4 \mu$s. In order to estimate the density of NV center, we measure the fluorescence saturation curve of NV center ensemble. From Figure A1(b), we infer that there are $\sim 100$ NV center in our confocal microscopy spot and the typical size of spot is $300 \times 300 \times 600 \text{nm}^3$. Then we can get the density of NV center is $[\text{NV}]e0.0063 \text{ppm}$. The density of [N] is about 2 ppm by analyzing of ESR signal with JES-FA200.

Appendix B: Analysis of the signal

The signal express as:

$$
\begin{align*}
S &= \text{Tr}\left[ P e^{-i\delta_{f}/h} R(\pi) e^{-i\delta_{f}/h} R(\frac{\pi}{2}) \rho R(\frac{\pi}{2}) e^{i\delta_{f}/h} R(\pi) e^{i\delta_{f}/h} \right],
\end{align*}
$$

We can always parameterize the $R(\theta)$, $P$ and $\rho_{\text{NV}}$ with Pauli matrix of spin $1/2$ in this way:

$$
\begin{align*}
R(2\theta) &= \cos \theta I + b_y \sigma_x + b_z \sigma_y + b_z \sigma_z, \\
P &= \frac{1}{2} a_x \sigma_x + a_y \sigma_y + a_z \sigma_z, \\
\rho_{\text{NV}} &= \left( \frac{1}{2} + c_x \sigma_x + c_y \sigma_y + c_z \sigma_z \right).
\end{align*}
$$

The above coefficients can be determined by calculation of matrix trace. We have:

$$
\begin{align*}
s &= C + A_1 \text{Tr}[U_0(\tau) \rho_0 U(\tau)] \cos(\delta + \gamma B_{\text{sat}} \tau) + A_2 \text{Tr}[U_0(\tau) \rho_0 U(\tau)] \sin(\delta + \gamma B_{\text{sat}} \tau) \\
&+ B_1 \text{Tr}[U_0(\tau) \rho_0 U_1(\tau)] \cos(\delta - \gamma B_{\text{sat}} \tau) + B_2 \text{Tr}[U_0(\tau) \rho_0 U_1(\tau)] \sin(\delta - \gamma B_{\text{sat}} \tau) \\
&+ D_1 \text{Tr}[U_0(2\tau) \rho_0 U(2\tau)] \cos 2\delta \tau + D_2 \text{Tr}[U_0(2\tau) \rho_0 U(2\tau)] \sin 2\delta \tau \\
&+ E_1 \text{Tr}[U_0(\tau) U(\tau) \rho_0 U_0(\tau) U_1(\tau)] \cos 2\gamma B_{\text{sat}} \tau + E_2 \text{Tr}[U_0(\tau) U(\tau) \rho_0 U_0(\tau) U_1(\tau)] \sin 2\gamma B_{\text{sat}} \tau
\end{align*}
$$

with

$$
\begin{align*}
C &= \frac{1}{2} - 2a_x c_x \cos^2 \theta - 2a_y b_y c_y - 2a_z b_z c_z + 2a_x b_x c_x, \\
A_1 &= -4a_x b_y c_y \cos \theta + 4a_y b_y c_y \cos \theta - 4a_x b_z c_z - 4a_y b_z c_z, \\
A_2 &= -4a_x b_y c_y \cos \theta - 4a_y b_y c_y \cos \theta + 4a_x b_z c_z + 4a_y b_z c_z, \\
B_1 &= 4a_x b_y c_y \cos \theta - 4a_y b_y c_y \cos \theta - 4a_x b_z c_z - 4a_y b_z c_z, \\
B_2 &= 4a_y b_y c_y \cos \theta + 4a_y b_y c_y \cos \theta + 4a_x b_z c_z + 4a_y b_z c_z.
\end{align*}
$$

Figure A1. (a) Corresponding magnetic field sensitivities with the power of 532 nm laser. (b) The fluorescence saturation curve versus excitation power.
\begin{align*}
B_z &= -4i a^2 c \cos \theta - 4i a b c \cos \theta - 4a^2 b c,
\end{align*}
\begin{align*}
D_z &= 2a^2 c \cos^2 \theta + 2a b c \cos^2 \theta - 4i a b c \cos \theta + 4i a b c \cos \theta - 2a b c - 2a b c,
\end{align*}
\begin{align*}
D_i &= 2a^2 c \cos^2 \theta - 2a b c \cos^2 \theta + 4i a b c \cos \theta + 4i a b c \cos \theta - 2a b c + 2a b c,
\end{align*}
\begin{align*}
E_z &= -4a^2 b c - 4a b c - 2a b c - 2a b c + 2a b c + 2a b c,
\end{align*}
\begin{align*}
E_i &= 4a^2 b c - 4a b c - 2a b c + 2a b c - 2a b c - 2a b c - 2a b c.
\end{align*}

The term $Tr[U_z(\tau) \rho_z U_z(\tau)]$, $Tr[U_z(2\tau) \rho_z U_z(2\tau)]$ is relative to the error operation of DD method ($\pi$ pulse). At last, we employ the cluster-correlation expansion (CCE) method to calculate above terms such as $Tr[U_z(\tau) \rho_z U_z(\tau)]$, $Tr[U_z(2\tau) \rho_z U_z(2\tau)]$, $Tr[U_z(\tau) U_z(\tau) \rho_z U_z(\tau) U_z(\tau)]$ and get results in main text.