All-optical magnetic resonance of high spectral resolution using a nitrogen-vacancy spin in diamond

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Abstract
We propose an all-optical scheme to prolong the quantum coherence of a negatively charged nitrogen-vacancy (NV) center in diamond at cryogenic temperatures. Optical control of the NV spin suppresses energy fluctuations of the $^3A_2$ ground states and forms an energy gap protected subspace. By optical control, the spectral linewidth of magnetic resonance is much narrower and the measurement of the frequencies of magnetic field sources has higher resolution. The optical control also improves the sensitivity of the magnetic field detection and can provide measurement of the directions of signal sources.

Keywords: magnetic resonance spectroscopy, optical control, nitrogen-vacancy center

1. Introduction
Magnetic resonance is currently one of the most important tools in many areas of science and technology, including analytical chemistry, materials science, structural biology, neuroscience, and medicine [1]. However, the sensitivity of conventional techniques is restricted to large spin...
ensembles, which currently limits spatial resolution to the micrometer scale [2]. Recently considerable attention has focused on the application of negatively charged nitrogen-vacancy (NV) centers in diamond as an atomic-sized magnetic field sensor [3–13], where the NV centers are initialized and read out by optical fields [14, 15]. To suppress the effect of environmental noise and to obtain magnetic resonance signals of the sensing targets, microwaves are used to implement pulsed [16–20] and continuous [21, 22] dynamical decoupling control. The magnetic resonance signals represent important pieces of information to identify target spins and their relative positions of high spatial resolution [6, 7, 9–12]. The magnetic resonance frequencies are determined by the pulse rates of pulsed dynamical decoupling [6, 7, 9] or the Rabi frequencies of continuous dynamical decoupling [10–12]. Increasing the pulse rates in the case of pulsed dynamical decoupling and their Rabi frequencies in the case of continuous dynamical decoupling beyond the GHz regime is highly challenging. The requirement of realizing microwaves (which have wavelengths of centimeters) imposes limitations on the setup and individual microwave control on NV centers is difficult. Hence there has been a major effort to desirable to develop methods to overcome these shortcomings. All-optical control was recently shown to be possible [23], and an all-optical scheme for sensing the amplitudes of magnetic fields was demonstrated by electromagnetically induced transparency in an NV ensemble [24]. However to date, there are no all-optical methods to measure the frequencies of the magnetic fields that provide rich magnetic resonance information about the signal sources.

It is well-know that by optical driving fields one can realize a dark state which is decoupled from the excited states [25]. The fundamental principle underlying dark states is destructive interference which can be perturbed by external dephasing noise. If the dephasing noise has a finite bandwidth and when the dark state is separated in energy from all other bright states by more than this bandwidth, the sensitivity of the dark state to dephasing noise can be reduced considerably. Indeed, a qubit encoded in a dark state and an auxiliary stable state has been shown to be resilient to low frequency environmental noise and amplitude fluctuations in the driving field [26, 27]. The resulting enhanced coherence time enables longer coherent interaction with sensing targets and better sensing signals.

Here, we demonstrate the working principle by considering a negatively charged NV center at cryogenic temperatures. Specifically, we propose an all-optical magnetic resonance scheme using a negatively charged NV center to measure the frequencies of magnetic fields (see figure 1(a)). For the case that the magnetic resonance frequencies are determined by the pulse rates of pulsed dynamical decoupling [6, 7, 9] or the Rabi frequencies of continuous dynamical decoupling [10–12], errors in the dynamical decoupling control can broaden the resonant signal peaks. In contrast, in our scheme fluctuations in optical control do not broaden the resonant signal peaks, and the frequency of magnetic resonance is determined by the energy gap of the $^3A_2$ ground sublevels, which can easily extend the sensing frequencies to the GHz range. The optical control of the NV center suppresses the energy fluctuations of the $^3A_2$ ground sublevels and significantly extends the coherence times of the NV centers. Since the magnetic resonance linewidth broadening by dephasing is eliminated through the optical control, magnetic resonance of high spectral resolution with NV centers becomes possible. Higher spectral resolution can also improve measurement of the spatial positions of target spins when the schemes utilize magnetic resonance signals [6, 7, 12]. The all-optical magnetic resonance scheme may also have applications in solid-state GHz frequency standards and in all-optical quantum information processing with NV centers.
2. A negatively charged NV center under optical control

In applications of negatively charged NV centers in quantum technologies, it is important to prolong the quantum coherence of the triplet spin ground states $|\pm 1g\rangle$ and $|1g0\rangle$, where $|1g0\rangle$ refers to the orbital state with zero orbital angular momentum projections along the NV axis. The relaxation time of an NV spin can approach 200 s at low temperatures (10 K) [28], whereas the dephasing time is relatively short, with typical values for the inhomogeneous dephasing time $T_{2*}$ of 0.5–5 μs [14, 15]. With a large energy difference between $|0g\rangle$ and $|1g\rangle$, dephasing is the main source of decoherence and limits the overall decoherence time.

For simplicity, we model the dephasing by magnetic field fluctuations $\beta(t)$ on the NV axis (along $z$ direction), which couple to the NV spins through the Zeeman interaction ($\hbar = 1$)

$$H_{\text{dep}} = \beta(t) S_z,$$

with the spin operator $S_z = |+1g\rangle\langle +1g| - |-1g\rangle\langle -1g|$. We assume that $\beta(t)$ has zero-mean $\overline{\beta(t)} = 0$, where the overline denotes ensemble averaging. The random field fluctuations $\beta(t)$ induce broadening of the states $|\pm 1g\rangle$. An initial state of the center spin $|\Psi(0)\rangle = \sum_{k=\pm,0} a_k |k_g\rangle$ driven by $H_{\text{dep}}$ will evolve to $|\Psi(t)\rangle = a_{-1} e^{i\varphi(t)} |1g\rangle + a_0 |0g\rangle + a_+ e^{-i\varphi(t)} |+1g\rangle$, where the accumulated random phase $\varphi(t) = \int_0^t \beta(\tau)d\tau$. The coherence between $|0g\rangle$ and $|\pm 1g\rangle$ is described by the average of the relative random phase factor $L_{0,\pm} = \overline{e^{i\varphi(t)}}$, which vanishes when the random phase is large. For Gaussian noise, $L_{0,\pm} = \exp\left[-\frac{1}{2} \sqrt{\varphi(t)\varphi(t)} \right]$.

To suppress the dephasing using only optical control, we use two laser fields resonantly coupling the triplet ground states $|\pm 1g\rangle$ to the $^3E$ excited state $|A_2\rangle = c_+ |E_-\rangle |+1\rangle + c_- |E_+\rangle |-1\rangle$, with $|c_+|^2 + |c_-|^2 = 1$ (see figure 1). The lasers also couple the states $|\pm 1g\rangle$ and $|A_1\rangle = c_+^* |E_-\rangle |+1\rangle - c_-^* |E_+\rangle |-1\rangle$ but with a large detuning $\delta$, which is the energy gap between the states $|A_2\rangle$ and $|A_1\rangle$. The optical transitions between $^3A_2$ and $^3E$ are spin conserving [29–31]. The state properties of the NV centers, such as the parameters $c_+$ and $c_-$, depend on electric, magnetic, and strain fields. The effective Hamiltonian to obtain the eigenstates and
eigenenergies of the $^3\text{E}$ levels at low temperatures can be found in the review paper [14]. To have well-resolved excited states, we put the NV center at cryogenic temperatures ($\leq 10$ K). Using $|E_-\rangle|1 + 1\rangle = c_+^*|A_2\rangle + c_-|A_1\rangle$ and $|E_+\rangle|1 - 1\rangle = c_-^*|A_2\rangle - c_+|A_1\rangle$, we have the Hamiltonian under constant optical control

$$H_0 = (\Omega_+ e^{i\phi_+} e^{i\omega_+ t} \left[ c_+^* |A_2\rangle + c_- |A_1\rangle \right] \langle +1_g | + \text{h.c.} ) + (\Omega_- e^{i\phi_-} e^{i\omega_- t} \left[ c_-^* |A_2\rangle - c_+ |A_1\rangle \right] \langle -1_g | + \text{h.c.} ) - \delta |A_1\rangle \langle A_1 | + \sum_{k_g = \pm 1_g, 0_g} E_{k_g} \langle k_g | k_g \rangle + H_{\text{dep}} + H_{\text{sig}},$$

(3)

where $\Omega_\pm$ are the Rabi frequencies and $E_{k_g}$ are the energies of the ground states. $\phi_\pm$ and $\omega_\pm$ are the phases and frequencies of the lasers, respectively. We also include an interaction Hamiltonian $H_{\text{sig}}$ for possible signal sources. The energy of $|A_2\rangle$ is set as the reference energy. The laser fields resonantly drive the transitions between $| \pm 1_g \rangle$ and $|A_2\rangle$ with the laser detuning $\Delta_{\pm 1_g} = E_{\pm 1_g} - \omega_\pm = 0$. In the rotating frame of $e^{-iH_{\text{sig}}t}$ with

$$H_g = \sum_{k_g = \pm 1_g} \left( E_{k_g} - \Delta_{k_g} \right) \langle k_g | k_g \rangle + E_{0_g} \langle 0_g | 0_g \rangle,$$

(4)

the system Hamiltonian reads

$$H = H_L + H_{\text{dep}} + \tilde{H}_{\text{sig}},$$

(5)

where

$$H_L = (\Omega_+ e^{i\phi_+} \left[ c_+^* |A_2\rangle + c_- |A_1\rangle \right] \langle +1_g | + \text{h.c.} ) - \delta |A_1\rangle \langle A_1 | + (\Omega_- e^{i\phi_-} \left[ c_-^* |A_2\rangle - c_+ |A_1\rangle \right] \langle -1_g | + \text{h.c.} ),$$

(6)

$$\tilde{H}_{\text{sig}} = e^{iH_{\text{sig}}t} H_{\text{sig}} e^{-iH_{\text{sig}}t}.$$  

(7)

For accurate numerical simulations, we model the NV spin with six levels: three ground states $| \pm 1_g \rangle$ and $|0_g \rangle$, the two excited states $|A_1\rangle$ and $|A_2\rangle$, and a singlet state $|s\rangle$ to describe the intersystem crossing transitions. The dynamics of the NV center spin described by a density matrix $\rho(t)$ is governed by the Lindblad master equation [32]

$$\frac{d}{dt} \rho = -i [H, \rho] + \sum_{\alpha, \beta} \gamma_{\beta \alpha} \left( \sigma_{\beta \alpha} \rho \sigma_{\alpha \beta} - \frac{1}{2} \rho \sigma_{\alpha \beta} - \frac{1}{2} \sigma_{\alpha \beta} \rho \right),$$

(8)

where the Lindblad operators $\sigma_{\beta \alpha} \equiv |\beta\rangle \langle \alpha |$, $\gamma_{\beta \alpha}$ are the decay rates, and the Hamiltonian $H$ is given by equation (5).

### 3. Noise suppression by optical control

To illustrate the fundamental idea of noise suppression by optical control, we consider a simplified model without contributions from spontaneous decay (taken into account in detailed numerical simulations to present subsequently). When the energy gap $\delta \gg \Omega_\pm$, the coupling to $|A_1\rangle$ is negligible in equation (5), and we have the $A$-type Hamiltonian by dropping out terms
related to the state $|A_1\rangle$

$$H_A = H_{L2} + H_{dep} + \tilde{H}_{sig},$$

where

$$H_{L2} = \Omega |A_2\rangle \langle b_g| + \text{h.c.},$$

with the effective Rabi frequency

$$\Omega \equiv \sqrt{\Omega_2^2 |c_+|^2 + \Omega_1^2 |c_-|^2},$$

and the bright state

$$|b_g\rangle \equiv \frac{1}{\Omega} \left( c_+ \Omega_+ e^{-i\phi_c} |1_g\rangle + c_- \Omega_- e^{-i\phi_c} |1_g\rangle \right).$$

The laser driving fields form a $\Lambda$ system with an excited state $|A_2\rangle$ and two ground states $|\pm 1_g\rangle$ (see figure 1). The dark state decoupled from the laser is

$$|d_g\rangle \equiv \frac{e^{i\phi_d}}{\Omega} \left( c_+ \Omega_+ e^{-i\phi_d} |1_g\rangle - c_- \Omega_- e^{-i\phi_d} |1_g\rangle \right),$$

with a global phase $\phi_d = \text{arg}(c_+ c_-)$. The laser Hamiltonian $H_{L2}$ has the eigenstates $|l_d\rangle, |l_0\rangle$, and $|l_{\pm}\rangle = \frac{1}{\sqrt{2}} (|b_g\rangle \pm |A_2\rangle)$ with energies $E_{l_{\pm}} = \pm \Omega$. The subspace spanned by the two states $|l_d\rangle$ and $|l_0\rangle$ are separated from the other eigenstates $|l_{\pm}\rangle$ by the energy gaps $\pm \Omega$. Therefore the transitions from this subspace to $|l_{\pm}\rangle$ are suppressed by an energy penalty that is proportional to the Rabi frequency of the optical driving fields (see figure 1 (b) and [26]).

The spin operator $S_z$ in the basis of $|l_d\rangle$ and $|l_g\rangle$ reads

$$S_z = \frac{1}{\Omega^2} \left( 2 e^{-i\phi_d} c_+ \Omega_+ c_- |l_g\rangle \langle b_g| + \text{h.c.} \right) + \kappa \left( |b_g\rangle \langle b_g| - |d_g\rangle \langle d_g| \right),$$

where $\kappa = \frac{1}{\Omega^2} (|c_+ \Omega_+|^2 - |c_- \Omega_-|^2) \leq 1$. To suppress the dephasing of the NV spin using the energy penalty $\Omega$, we choose the laser fields that satisfy

$$|c_+ \Omega_+| = |c_- \Omega_-|.$$

Under such a condition, the spin operator $S_z$ becomes

$$S_z = |b_g\rangle \langle d_g| + \text{h.c.} = \frac{1}{\sqrt{2}} (|l_+\rangle + |l_-\rangle) \langle d_g| + \text{h.c.},$$

with

$$|b_g\rangle = e^{i\phi_d} \frac{1}{\sqrt{2}} \left[ |1_g\rangle + e^{i\phi_L} |1_g\rangle \right],$$

$$|d_g\rangle = e^{i\phi_d} \frac{1}{\sqrt{2}} \left[ |1_g\rangle - e^{i\phi_L} |1_g\rangle \right].$$
where $\varphi_b = \arg(c_+ \Omega_+ e^{-i \phi_b})$ and

$$\phi_L = \arg(\Omega_+ \Omega_- c_+^* c_-) + \phi_+ - \varphi_b,$$

(19)
is a tunable relative phase. With a relatively large $\Omega$, the spectral power density of the magnetic field fluctuations $\beta(t)$ at the frequencies around the energy gap $\Omega$ is negligible. The off-resonant fluctuations $\beta(t)$ cannot induce the transitions from $|d_g\rangle$ to $|l_\pm\rangle$, which are strongly suppressed by the energy penalty $\pm \Omega$ (see figure 1(b)). Because fluctuations in the effective Rabi frequency $\Omega$ only cause small changes in the magnitudes of the energy gap, our scheme is stable against the fluctuations of $\beta(t)$ [26], as long as the magnitudes of the energy gap are still much larger than the fluctuation frequencies of $\beta(t)$.

The coherence $L_{0,d_g}(t)$ can be simplified as

$$L_{0,d_g}(t) = \langle d_g \mid U(t) \mid d_g \rangle \langle 0_g \mid U_{d}^\dagger(t) \mid 0_g \rangle,$$

(20)

where $U(t) = T e^{-i \int_0^t H_{d} \, dt}$ with the time-ordering operator $T$. The coherence $L_{0,d_g}(t)$ decreases when the absolute value of $L_{0,d_g}(t)$ decreases.

With optical driving fields, $L_{0,d_g}(t) = \exp \left[ -\frac{1}{2} \left( \int_0^t \beta(t) \, dt \right)^2 \right]$ for Gaussian noise and vanishes when the random phase is large. From the expression of equation (23), we can see that when the noise is relatively slow compared to the frequency $\Omega$, the effect of the noise $\beta(t)$ is averaged out by the oscillating functions $\cos \Omega t$ and $\sin \Omega t$. Dynamical decoupling also uses similar modulation functions to average out unwanted noise [19–22]. A more detailed analysis in frequency domain is given in appendix A.

To demonstrate our scheme, we performed numerical simulations by implementing the master equation (8), including the spontaneous decay and dephasing. We used the parameters in the experimental paper [23] for the numerical simulation. The decay rate from the excited states $|A_1\rangle$ and $|A_2\rangle$ to the ground states $|\pm 1_g\rangle$ and $|0_g\rangle$ is $\gamma_{g,e} \approx 17 \text{ MHz}$; the rate for intersystem crossing from the excited states to the singlet $|s\rangle$ is $\gamma_{s,e} \approx 37 \text{ MHz}$; the inverse intersystem crossing rate from $|s\rangle$ to the ground states is $\gamma_{s,g} \approx 2.7 \text{ MHz}$. In the simulation, the noise fluctuations $\beta(t)$ were simulated by the Ornstein–Uhlenbeck process [33], which is Gaussian.
Generated by the Ornstein–Uhlenbeck processes, the expectation value of $\beta_t(z)$ is 
\[
\beta_t(z) = \tau - \beta t e^{z(t) 0 (0)};
\]
the two-point correlation $\beta'_{tt}(z) \approx -2 - 2 e^{z(t) 0 (0)}$, where $t_0$ is the starting time to generate an Ornstein–Uhlenbeck process and $\beta_c$ is a diffusion coefficient [34]. These quantities converge to stationary values after a time larger than the correlation time $\tau$. In the simulation, we chose $\beta = 10^{\mu s}$ for $\beta_c = 25$. We used $\tau = 25$ in the simulation [34].

The realizations of Ornstein–Uhlenbeck processes were generated by the exact simulation algorithm in [36] which requires the generation of a unit Gaussian random number $G$ at each time step. The algorithm is exact because the update algorithm equation (24) is valid for any finite time step $\Delta t$ [36]. We chose the value of the diffusion coefficient $c_\beta \approx 2(T^2_\tau 2)$ so that without optical driving fields $L_{0,\pm 1}(T^2_\tau) = e^{-1}$ at the dephasing time $T^2_\tau = 3 \mu s$.

To demonstrate the coherence protection by optical control, we prepared the quantum state in the superposition $|\Psi(0)\rangle = \frac{1}{\sqrt{2}} (|d_g\rangle + |l_0_g\rangle)$. With this initial state and using the master equation (8) in the simulations, the quantum coherence $L_{0,d_g}(t) = 2\langle d_g |p(t)|l_0_g\rangle$ describes $L_{0,d_g}(t)$ in the simplified model (9) without contributions from spontaneous decay). We had examined $|L_{0,d_g}(t)|$ as a function of time $t$ under optical driving fields of different Rabi frequencies $\Omega$. We found that there is an optimal working point of $\Omega$ for coherence protection. A plot of $|L_{0,d_g}(t)|$ as a function of $\Omega$ was shown in figure 2, where the coherence was measured at $t = 50 \mu s$. From simulations for other choices of $t$, we observed that the optimal $\Omega$ is independent of the measurement times $t$ (which were chosen to be larger than a few $\mu s$ for a clear inspection). The existence of an optimal $\Omega$ is a result of the leakage to the excited states. When the optical driving fields are turned on, the dephasing Hamiltonian $H_{\text{dep}}$ in equation (5) causes leakage to the excited state $|A_2\rangle$, via the successive transitions $|d_g\rangle \rightarrow |l_0_g\rangle \rightarrow |A_2\rangle$. By finding the instantaneous eigenstates, the population in the state $|A_2\rangle$ was estimated as

![Figure 2. Quantum coherence between states $|d_g\rangle$ and $|l_0\rangle$ at the moment $t = 50 \mu s$ as a function $\Omega$, for (a) $B_{z,\text{bias}} = 0$ and (b) $B_{z,\text{bias}} \approx 0.1 T$. The data were obtained by $4 \times 10^4$ runs of averaging.](attachment:image.png)
$R_{\Omega} \sim \left( \frac{\tilde{\beta} \Omega}{\Delta^2 + \tilde{\beta}^2} \right)^2$, where $\tilde{\beta}$ is the effective noise strength. The population leakage out of the subspace spanned by $|l_d\rangle$ and $|l_0\rangle$ reduces the quantum coherence $|L_{0,ci}(t)|$. This effect can be seen from the reduction of coherence when $\Omega$ increases from 0 to $\sim$1 MHz in figure 2. Large driving field amplitudes $\Omega$ lead to a recovery of the quantum coherence by suppressing the noise effects (see the results in figure 2 for 1 MHz $\leq \Omega \leq$ 7 MHz). However, increasing $\Omega$ also increases the coupling to the state $|A_1\rangle$ (see equation (6)), which degrades the approximated $\Lambda$-type system (see equation (9)) and leads to population leakage to $|A_1\rangle$. At zero static field, the energy gap $\delta \approx$ 2 GHz and the mixing coefficients $|c_\pm| = |c_-| = 0.178$ in equation (6), and therefore $|d_\Omega\rangle$ only directly couples to $|A_1\rangle$. For $\Omega \ll \delta$, the population to $|A_1\rangle$ state is $R_{\Omega} \sim (2\Omega/\delta)^2$, estimated by time-independent perturbation theory. Because of relatively large decay rates on the excited states $|A_2\rangle$ and $|A_1\rangle$, we expect an optimal $\Omega > 0$ in our scheme when the leakage $R_{\Omega} + R_{\Omega} \sim$ a minimum. In figure 2(a), the optimal control with $\Omega \sim$ 10 MHz gives the best coherence protection. We can increase $\delta$ by applying a static bias magnetic field $B_{z,bias}$ along the axis of the NV center (z direction). However, non-zero $B_{z,bias}$ also changes the mixing coefficients $c_\pm$. For example, $B_{z,bias} = 0.1$ T gives $\delta \approx 5.71$ GHz, $c_+ \approx 0.984$, and $c_- \approx 0.178$. When $|c_\pm| \neq |c_-|$, the dark state $|d\rangle$ can transit to both excited states $|A_1\rangle$ and $|A_2\rangle$, and $|L_{0,ci}(t)|$ is strongly reduced (see figure 2(b)). At $B_{z,bias} = 0.1$ T, the effective Rabi frequency $\Omega \sim$ 7 MHz yields the best coherence protection.

Having obtained the optimal driving amplitudes of $\Omega$, in figure 3 we plotted the quantum coherence as a function of time for cases of free induction decay ($\Omega = 0$) and with optical driving fields ($\Omega = 10$ MHz) at zero static fields. With an optical Rabi frequency $\Omega = 10$ MHz, the coherence is significantly prolonged and exceeds 50 $\mu$s, which is over 16-fold improvement in the coherence time $T_2^\ast \approx 3$ $\mu$s. Since the coherence can be preserved up to $\sim$50 $\mu$s and a longer evolution time provides better spectral resolution, in the following simulations, we will perform measurement at the time $t = 50$ $\mu$s.

Although the scheme is robust to the fluctuations of $\Omega$ [26], equation (15) implies that independent fluctuations $\delta \Omega_\pm(t)$ in the amplitudes of $\Omega_\pm$ change the bright and dark states in equations (17) and (18). To demonstrate that the scheme is not sensitive to independent fluctuations $\delta \Omega_\pm(t)$, we modelled $\delta \Omega_\pm(t)$ by Ornstein–Uhlenbeck processes. We selected a diffusion coefficient $c_\Omega = 2\delta_\Omega^2/\tau_\Omega$ with a correlation time $\tau_\Omega = 100$ $\mu$s and a variance of relative
fluctuations $\delta \Omega = \pm \Omega t \Omega$. The impact of independent intensity fluctuations on the coherence is shown in figure 4. The scheme still provides good performance even though the standard deviation of relative fluctuations $\delta \Omega$ reaches $\sim 0.02$.

4. High-resolution magnetic resonance by optical control

In magnetic resonance spectroscopy, when the energy gap, e.g., $\epsilon_{0,-1} \equiv E_0 - E_{-1}$ between $|0_g\rangle$ and $|1_g\rangle$, coincides with the frequency of the magnetic field, resonance transitions induce a signal peak in the frequency domain. The linewidth and depth of the peak determine the resolution and sensitivity of the spectroscopy. Because of the energy broadening induced by the random fluctuations $\beta(t)$, the minimum linewidth of the signal peak is limited by the deviation of the fluctuations $\sim \langle \beta(t)^2 \rangle^{1/2}$. When the dephasing (i.e., the effect of energy broadening) is suppressed, we achieve a narrower linewidth, and hence spectroscopy with higher accuracy.

Before giving the details, we summarize the steps of the sensing scheme. We first initialize the NV center in the state $|0_g\rangle$ by optical pumping. To start the sensing, we turn off the pumping lasers and turn on the driving fields to form the dark state. After some evolution time, we turn off the driving fields and measure the population at $|0_g\rangle$ by optical readout. The population at $|0_g\rangle$ gives the magnetic resonance signals.

We assume that the signal fields have negligible frequency components around the energy gap $\Omega$. The signal Hamiltonian in equation (9) is written as

$$\hat{H}_{\text{sig}} = \eta_{\text{sig}}(t) e^{iH_\Omega t} \left[ S_x \cos \theta_{\text{sig}} + S_y \sin \theta_{\text{sig}} \right] e^{-iH_\Omega t},$$

where $\eta_{\text{sig}}(t)$ is the magnetic signal field with zero mean $\eta_{\text{sig}}(t) = 0$ and $\theta_{\text{sig}}$ is the direction of the magnetic field in the $x$-$y$ plane normal to the NV axis. We initialize the NV center in the state $|0_g\rangle$ by optical pumping. The population that remains in the initial state $|0_g\rangle$ is approximately governed by the dynamics induced by $H_\lambda$ given by equation (9).
In the simulation, we generated single-frequency sources \( \eta \) with initial random phases \( \phi \) at each run of the simulation. To have the accurate resonance frequency \( \omega_{\text{Res}} \), we diagonalized the Hamiltonian \( H \) given by equation (6), as \( \omega_{\text{Res}} \) is the energy between \( |0_g \rangle \) and the state \( |d \rangle \) which is approximately \( |g \rangle \) and has a little mixing with the excited states \( |A_2 \rangle \) and \( |A_1 \rangle \). For \( \Omega \ll \delta, \omega_{\text{Res}} \approx \epsilon_{0,1} \), up to a correction \( \sim \Omega^2/\delta \). We consider the magnetic resonance signal where the difference between the signal frequency and the resonant frequency is relatively small, i.e., \( |(\omega - \omega_{\text{Res}})|/\omega_{\text{Res}} | \ll 1 \). This enables the application of a rotating wave approximation by neglecting oscillating terms with frequencies \( \sim 2\omega_{\text{Res}} \) in equation (25). The simulations used the master equation (8) with the Hamiltonian equation (5).

4.1. Measurement of signal frequencies

When there is no static bias field \( (B_z, \text{bias} = 0) \), \( \delta \approx 2 \text{ GHz} \), the states \( | \pm 1_g \rangle \) are degenerate, and

\[
\hat{H}_{\text{sig}} = \eta_{\text{sig}}(t) e^{-i\epsilon_{0,1} t} e^{i\theta_{\text{us}}} \frac{1}{\sqrt{2}} \left( |1_g \rangle + |1_g \rangle e^{i2\theta_{\text{us}}} \right) \langle 0_g | + \text{h.c.} \tag{27}
\]

By choosing the laser phase \( \phi_\perp = 2\theta_{\text{sig}} + \pi \), we achieve the largest sensitivity as the signal Hamiltonian \( \hat{H}_{\text{sig}} = \eta_{\text{sig}}(t) e^{-i\epsilon_{0,1} t} e^{i\theta_{\text{us}}} e^{-i\phi_\perp} |d_g \rangle \langle 0_g | + \text{h.c.} \) The states \( |d_g \rangle \) and \( |0_g \rangle \) are coherence protected. We obtain the magnetic resonance signal by tuning the resonant frequency. When the resonant frequency is tuned to the frequency of the signal fields, the state \( |0_g \rangle \) will transit to \( |d_g \rangle \) and the change of \( P_{|0_g \rangle}(t) \) gives the signal.

In figure 5, we plot the magnetic resonance signal at the evolution time \( t = 50 \mu s \). The laser phase is \( \phi_\perp = 2\theta_{\text{sig}} + \pi \), and the signal fields have an amplitude \( \eta_0 = 0.01 \text{ MHz} \). Without optical control, the signal fields with frequency \( \omega_{\text{Res}} \) lead to a maximum resonant population change \( \delta P_{|0_g \rangle} \approx 21\% \) with a linewidth \( \Delta\omega_{\text{FWHM}} \approx 0.2 \text{ MHz} \) (defined by the full width at half maximum); while with optical control \( \Omega = 10 \text{ MHz} \), the signal induces a much larger

\[
P_{|0_g \rangle}(t) = \left| \left\langle 0_g | e^{-i\int_0^t H_{\text{dr}} d\tau} |0_g \right\rangle \right|^2. \tag{26}
\]
population dip \(\delta P \approx 63\%\) with a much narrower linewidth \(\Delta \omega_{\text{FWHM}} \approx 0.02\) MHz, which is limited by the evolution time \(t\) (within the coherence time range).

To reduce the resonant frequency \(\omega_{\text{Res}}\) within the MHz range, we apply a static bias magnetic field along the axis of the NV center to narrow the energy gap \(\epsilon_{0,-1}\). When the magnetic field \(B_{z,\text{bias}} \approx 0.1\) T, \(\delta \approx 5.7\) GHz, the ground states \(|0_g\rangle\) and \(|1-1_g\rangle\) have an energy gap within the MHz range, and the energy gap between \(|0_g\rangle\) and \(|+1_g\rangle\) is large (\(\gtrsim 2.9\) GHz). For large energy gaps between \(|0_g\rangle\) and \(|+1_g\rangle\), the transition from \(|0_g\rangle\) to \(|+1_g\rangle\) induced by signal fields is negligible and we have

\[
\hat{H}_{\text{sig}} \approx \eta_{\text{sig}}(t) \frac{1}{2} e^{-i\epsilon_{0,-1}t} e^{i(\theta_{\text{sig}} - \phi - \phi_L)} \left(|b_g\rangle - |d_g\rangle\right)\langle 0_g \rangle + \text{h.c.} \tag{28}
\]

Under optical control with a large \(\Omega\), we can ensure that the spectral density of \(\beta(t)\) around \(\Omega\) and \(\eta_{\text{sig}}(t)\) around \(\epsilon_{0,-1} \pm \Omega\) is negligible. The transitions between \(|0_g\rangle\) and \(|b_g\rangle\) are suppressed, and we get

\[
\hat{H}_{\text{sig}} \approx -\eta_{\text{sig}}(t) \frac{1}{2} e^{-i\epsilon_{0,-1}t} e^{i(\theta_{\text{sig}} - \phi - \phi_L)} |d_g\rangle \langle 0_g | + \text{h.c.} \tag{29}
\]

Therefore, when the field \(\eta_{\text{sig}}(t)\) is on resonant with the transition frequency around \(\omega_{\text{Res}} \approx \epsilon_{0,-1}\), the population of \(|0_g\rangle\) decreases. In this way, the Fourier components of the signal source \(\eta_{\text{sig}}(t)\) can be measured in high resolution. In figure 6, \(\eta_0 = 0.02\) MHz, \(B_{z,\text{bias}} \approx 0.1\) T, and the evolution time \(t = 50\) \(\mu\)s. Without optical control, the signal fields lead to a maximum population dip \(\delta P_{0_g} \approx 30\%\) with a linewidth \(\Delta \omega_{\text{FWHM}} \approx 0.2\) MHz; while with optical control \(\Omega = 7\) MHz, the population has a larger peak \(\delta P_{0_g} \approx 42\%\) with a much narrower linewidth \(\Delta \omega_{\text{FWHM}} \approx 0.02\) MHz limited by finite evolution time.

In figures 5 and 6, we also plot the magnetic resonance signals for optical control with independent driving fluctuations \(\delta \Omega_\pm(t)\). It can be seen that an experimentally reachable standard deviation of relative fluctuations \(\delta_\Omega = 0.005\) only induces tiny changes in the magnetic resonance signals.
4.2. Measurement of the directions of signal sources

For the case of \((B_z,\text{bias}) = 0\), the signal Hamiltonian equation (27) depends on the direction \(\theta_{\text{sig}}\) of the signal source. Note that the transition from \(|0_g\rangle\) to \(|b_g\rangle\) is suppressed when there is optical control. If \(\phi_L = 2\theta_{\text{sig}}\) in equation (27), the effect of the signal Hamiltonian

\[
\hat{H}_{\text{sig}} = \eta_{\text{sig}}(t)e^{-i\omega_0 t - i\theta_0}e^{-i\phi_\theta}\hat{b}_g\langle 0_g| + \text{h.c.} \quad \text{is suppressed by the energy gap between } |0_g\rangle \text{ and } |b_g\rangle \text{ states, and the population change } \Delta R_{b_g} \text{ is small. We have shown that the signal is large when the laser phase } \phi_L = 2\theta_{\text{sig}} + \pi. \text{ We use this phase dependence to determine the direction } \theta_{\text{sig}} \text{ of the signal field.}
\]

In figure 7, we applied a resonant signal field at the frequency \(\omega_s = \omega_{\text{Res}}\) and tuned the laser phase \(\phi_L = 2\theta + \pi\) with different angles \(\theta\). The control strength \(\Omega = 10\ \text{MHz}\) and the amplitude of the signal field \(\eta_0 = 0.01\ \text{MHz}\). From the lowest point in figure 7, we can infer the direction \(\theta_{\text{sig}}\) of the signal source in the \(x-y\) plane. For the case without optical control, \(x\) and \(y\) directions are equivalent and it is obvious that we cannot infer the direction of the signal source in the \(x-y\) plane. Note that if the laser phase \(\phi_L\) has a small deviation in the region \(2(\theta_{\text{sig}} - 0.1\pi) + \pi \lesssim \phi_L \lesssim 2(\theta_{\text{sig}} + 0.1\pi) + \pi\), the signal \(\Delta R_{b_g}\) is also large with little change (\(\approx 1\%\)). Therefore, our scheme is robust to fluctuations of laser phase.

4.3. Sensitivity enhanced by optical control

Note that in figures 5 and 6, the signal peaks with optical control are more pronounced. This implies that the sensitivity is also improved when the dephasing noise is suppressed by the optical control. In figure 8, we plot the one-trial sensitivity [36]

\[
\delta P|\phi_s\rangle_{\text{min}} = \Delta P|\phi_s\rangle \left/ \left| \frac{\partial P|\phi_s\rangle}{\partial \eta_0} \right| \right. \quad \text{(30)}
\]

for the cases of the bias magnetic field \(B_z,\text{bias} \approx 0\) and \(B_z,\text{bias} \approx 0.1\text{T}\), respectively. The parameters are the same as those for the optical sensing in figures 5 and 6 with \(\omega_s = \omega_{\text{Res}}\). Here \(\Delta R_{b_g} = \sqrt{\left< \hat{P}_{b_g}(1 - \hat{P}_{b_g}) \right>}\) is the standard deviation of the population \(\hat{P}_{b_g}\) in one measurement. Averaging the data by repeating the measurement \(N\) times improves the sensitivity by a factor of \(\alpha_r = 1/\sqrt{N}\). If we perform the experiment for a given time \(T_{\text{all}}\), we have \(N = T_{\text{all}}/(T_{\text{init}} + t)\), where \(T_{\text{init}}\) is the time for both initialization and readout and \(t\) is the evolution time of the NV spin. With a large bias field, the transition from \(|0_g\rangle\) to \(|1 + 1_g\rangle\) is suppressed by the energy gap between \(|0_g\rangle\) and \(|1 + 1_g\rangle\) (see equation (28)). Therefore the effective signal strength is reduced and the sensitivity with optical control is reduced in figure 8 at a short sensing time \(t \lesssim 10\ \mu s\). At a longer sensing time, the benefits from decoherence suppression by optical control manifest and the sensitivity is improved. At zero bias field, the transition from \(|0_g\rangle\) to \(|1 + 1_g\rangle\) is kept under optical control, and the one-trial sensitivity is improved for a wide range of times \(t \lesssim 45\ \mu s\) in the figure.

With the chosen values of \(\eta_0\) in figure 8, we can see that the case of optical control has significantly lower sensitivity around \(t = 50\ \mu s\) (compared with the high performance around \(t \approx 30\ \mu s\)). To analyze the reduction of the one-trial sensitivity, we simplified the Lindblad master equation by using a Hamiltonian \(\eta_0|d_g\rangle\langle 0_g| + \text{h.c.}\) and used \(\gamma_b\) to describe an effective transition rate from the state \(|d_g\rangle\) to the states out of the subspace spanned by \(|d_g\rangle\) and \(|0_g\rangle\). The effective signal amplitude \(\eta_0 \approx \eta_0\) for the case of \(B_z,\text{bias} = 0\), and \(\eta_0 \approx \eta_0/2\) for \(B_z,\text{bias} \approx 0.1\text{T}\). By setting the leakage rate \(\gamma_b = 0\) in this simplified model, we obtained the idealized sensitivity.
 δR_{\theta_j},min \approx 1/(4\pi t) \text{ (MHz)}, which does not diverge at t > 0. For small \( \gamma_b \) and \( \eta_0 \) in the simplified model, we obtained an approximated population \( R_{\theta_j}(t) \approx \cos^2(2\eta_0 t)e^{-\eta_0 t} \) and found a divergence of sensitivity \( \delta R_{\theta_j},\text{min} \) when the signal phase is around \( 2\pi \eta_0 t = n\pi \) \( (n = 1, 2, \ldots) \). That is, with leakage, the sensitivity becomes significantly low around \( t = n/(2\eta_0) \) \( (n = 1, 2, \ldots) \). Also note that in figure 8 the sensitivity without optical control does not diverge at \( t = 1/(2\eta_0) = 50 \mu s \). Therefore, with optical control, the low sensitivity around \( t=50 \mu s \) in figure 8 is caused by the population leakage out of the subspace spanned by the sensing levels \( |d_{g}\rangle \) and \( |0_{g}\rangle \). In figure 8, we chose the values of \( \eta_0 \) to show the effect of population leakage on the sensitivity, and these values of \( \eta_0 \) were also used in figures 5 and 6, where relatively larger \( \eta_0 \) provided more pronounced resonance signals. The sensitivity at \( t = 50 \mu s \) can be improved if the strength of the signal sources \( \eta_0 \) is smaller.

**Figure 7.** The on-resonant (\( \omega_s = \omega_{Res} \)) magnetic signal (the change of the population in \( |0_{g}\rangle \)) as a function of the angle difference \( \theta - \theta_{\text{mag}} \) at the moment \( t = 50 \mu s \). The parameters \( B_{z,\text{bias}} \approx 0, \ \Omega = 10 \text{ MHz}, \ \text{and} \ \eta_0 = 0.01 \text{ MHz}. \) The data were obtained by \( 5 \times 10^3 \) runs of averaging.

**Figure 8.** The one-trial sensitivity of the magnetic resonance signal at the resonant frequency \( \omega_s = \omega_{Res} \) for the cases of without optical control (blue dots) and with optical control (red squares). (a) \( B_{z,\text{bias}} = 0, \ \eta_0 = 0.01 \text{ MHz}, \ \text{and} \ \Omega = 10 \text{ MHz}; \) (b) \( B_{z,\text{bias}} \approx 0.1 \text{ T}, \ \eta_0 = 0.02 \text{ MHz}, \ \text{and} \ \Omega = 7 \text{ MHz}. \) The data were obtained by \( 10^5 \) runs of averaging.
5. Discussion and conclusion

We have proposed an all-optical scheme to prolong the quantum coherence of a negatively charged NV center in diamond. With the quantum coherence extended and the energy fluctuations in the $^3A_2$ ground sublevels suppressed by the optical driving fields, we have achieved magnetic resonance with much narrower spectral linewidth and higher detection sensitivity. Unlike magnetic resonance by pulse sequences or more generally by dynamical decoupling, in our scheme driving field fluctuations do not broaden the resonant signal peaks. The sensing frequency by optical control is determined by the energy gap of the NV $^3A_2$ ground sublevels, which can easily reach the GHz range and enables stable GHz frequency standards in solids. At zero field, the magnetic resonance spectrum also enables measurement of the direction of signal sources in the plane perpendicular to the NV symmetry axis. The performance of the all-optical scheme has been confirmed by numerical simulations, by selecting the driving amplitudes within a range where the transitions to the off-resonant excited state are small and the $\Lambda$-type optical transition is a good approximation. In order to use the proposed scheme, the NV center is put at cryogenic temperatures ($\lesssim 10$ K) so that the linewidth of optical transitions is small. While this may be considered as a drawback compared to the dynamical decoupling schemes by microwaves [6, 7, 9–12], in which the NV centers can work at room temperatures, we would like to stress that our method is general and is applicable to other systems where a $\Lambda$-type optical transition can be formed. Furthermore, a wide variety of sensing tasks, e.g., of magnetism or superconductivity in solid state naturally take place at low temperatures and therefore provide a wide field of applicability of our proposed sensing scheme.

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Appendix A. Decoherence function in the frequency domain

Here we outline the decoherence suppression by optical control in the frequency domain. For simplicity, we consider a simplified model without contributions from spontaneous decay and examine the effect of $\tilde{\rho}(t)$ up to the second order

$$L_{0,d}(t) = 1 - \frac{1}{2} \int_0^t dt_1 \int_0^{t_1} dt_2 \tilde{\rho}(t_1) \tilde{\rho}(t_2) M(t_1, t_2),$$

(A.1)

where the modulation function

$$M(t_1, t_2) = \cos \Omega t_1 \cos \Omega t_2 + \sin \Omega t_1 \sin \Omega t_2,$$

(A.2)

$$= \cos [\Omega (t_1 - t_2)].$$

(A.3)
We assume stationary noise; i.e., noise with time translation symmetry, $eta(t_2) - eta(t_1) = \beta(t_2 - t_1)$ and $\beta(t_1) - \beta(t_2)$. Using

$$L_{0,d_0}(t) = 1 - \frac{1}{2} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} S_\beta(\omega) \tilde{M}(\omega),$$

(A.4)

in terms of the spectral power density

$$S_\beta(\omega) = \int_{-\infty}^{\infty} dt \beta(t) \beta(t) e^{i\omega t},$$

(A.5)

and the filter function

$$\tilde{M}(\omega) = \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 e^{-i\omega(t_1-t_2)} M(t_1, t_2).$$

(A.6)

Without optical control, i.e., $\Omega = 0$, the function $\tilde{M}(\omega) = 4 \sin^2 \left( \frac{\omega t}{2} \right) / \omega^2$ cannot filter out low-frequency fluctuations, which are dominant sources of decoherence. With large optical driving fields, the filter function $\tilde{M}(\omega) \approx C_{\omega,\Omega} \sin^2 \left( \frac{2\omega t}{2} \right) / \Delta^2_{\omega,\Omega}$ with $C_{\omega,\Omega} = 8\omega t / (\omega + \Omega)^2$ has a power-law decay with the deviation $\Delta_{\omega,\Omega} = |\omega - \Omega|$, and the low frequency fluctuations are filtered out for large $\Omega$. In this simplified model, when the spectral power density around the frequency $\Omega$ is negligible, $|L_{0,d_0}(t)| \approx 1$ and the decoherence is strongly suppressed.

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