# **Ultrasensitive diamond magnetometry using optimal dynamic decoupling**

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Magnetometry techniques based on nitrogen-vacancy (NV) defects in diamond have received much attention of late as a means to probe nanoscale magnetic environments. The sensitivity of a single NV magnetometer is primarily determined by the transverse spin-relaxation time, *T*2. Current approaches to improving the sensitivity employ crystals with a high NV density at the cost of spatial resolution or extend  $T<sub>2</sub>$  via the manufacture of novel isotopically pure diamond crystals. We adopt a complementary approach in which optimal dynamic decoupling techniques extend coherence times out to the self-correlation time of the spin bath. This suggests single spin, room-temperature magnetometer sensitivities as low as 5 pT Hz<sup>-1/2</sup> may be possible with current technology.

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# **I. INTRODUCTION**

Room-temperature single spin magnetometry using the nitrogen-vacancy (NV) center in diamond has the potential to revolutionize nanoscale imaging through fundamentally new detection modes. $1-8$  $1-8$  Proposals to image nanoscale environments exhibiting static (dc) and oscillatory (ac) magnetic fields using NV systems $2,3$  $2,3$  have since been demonstrated experimentally. $4-6$  $4-6$  By exploiting measured changes in quan-tum decoherence<sup>7[,8](#page-4-2)</sup> these techniques have been extended to include more general classes of randomly fluctuating fluctuating current (FC)] magnetic fields with comparable sensitivity to the ac case. $8$  One prominent reason such roomtemperature single spin-detection techniques are of interest is because of the potential to develop fundamentally new imaging modes for biological systems with nanometer resolution.<sup>9</sup>

The sensitivity of an NV magnetometer is governed by the transverse spin-relaxation (dephasing) time  $T_2$ , which for ac detection using isotopically pure diamond has been demonstrated at 4  $nT Hz^{-1/2.6}$  $nT Hz^{-1/2.6}$  $nT Hz^{-1/2.6}$  Many important detection problems, particularly those related to biology, $9$  stand to gain significant improvements with increased coherence times. In this paper we show how optimal dynamic decoupling techniques<sup>10</sup> can be exploited to increase sensitivities by over 2 orders of magnitude. With sensitivities in the pT  $Hz^{-1/2}$ regime, and nanoscale spatial resolution, the ultrasensitive NV magnetometer proposed here may have profound implications for nanobioimaging and sensing.

It is well known that coherence times may be improved with the use of concatenated  $(CDD)$ ,<sup>[11](#page-4-10)</sup> random, and periodic dynamic decoupling schemes.<sup>12,[13](#page-4-12)</sup> The more recent Uhrig DD (UDD) scheme was shown to be optimal for decoupling a spin qubit from a bosonic bath, $10$  and has since been shown to be optimal for all systems in which dephasing is the dominant decoherence channel[.14](#page-4-13) This optimality stems from the required UDD resources scaling linearly with the order to which the environmental effects are suppressed. However, UDD is unable to suppress longitudinal relaxation, whereas CDD can. The use of UDD is ideally suited to the NV center

owing to long relaxation times  $[T_1 > 1 \text{ s (Ref. 6)}]$  $[T_1 > 1 \text{ s (Ref. 6)}]$  $[T_1 > 1 \text{ s (Ref. 6)}]$ , even at room temperature. The large Debye temperature allows for negligible decoherence via phonon excitation of the crystal lattice and a large zero-field splitting (2.88 GHz) of the ground-state magnetic sublevels prevents longitudinal spinspin relaxation. Hence longitudinal relaxation may be neglected, ensuring UDD is the optimal decoupling method.

Both ac and FC magnetometry schemes are based upon a spin-echo microwave control sequence in which a  $\pi$  pulse is used to flip the qubit at the halfway point of its evolution [Fig.  $1(c)$  $1(c)$ ], suppressing any quasistatic effects of the spin bath. The ac scheme is concerned with detection of fields of the form  $b_{\text{ac}} \sin(\nu t)$ , where the  $\pi$  pulse coincides with *t*  $=\pi/\nu$ , ensuring a nonzero integral of the field trace, and hence maximal phase shift of the NV spin. In this paper, we incorporate UDD into ac and FC magnetometry schemes and

<span id="page-0-0"></span>

FIG. 1. (Color online) (a) NV-center diamond lattice defect. (b) NV spin detection through optical excitation and emission cycle. Magnetic sublevels  $m_s = 0$  and  $m_s = \pm 1$  are split by a *D* =2.88 GHz crystal field. Degeneracy between the  $m<sub>s</sub> = \pm 1$  sublevels is lifted by a Zeeman shift,  $\delta\omega$ . Application of 532 nm green light induces a spin-dependent photoluminescence and pumping into the  $m_s = 0$  ground state. (c) Examples of controlled-ac fields (solid) as seen by the NV center (dashed) in the presence of the first, second, and fifth UDD sequence. Negative regions of the ac trace are mapped to positive, ensuring maximal phase accumulation of the NV spin. Slow FC fields, such as the surrounding nuclear spin bath, will be suppressed, permitting the detection of ac and fast FC fields with greater sensitivity.

show that the sensitivity of a single NV center magnetometer may be as low as 5 pT  $Hz^{-1/2}$  with the use of existing technology.

Recently it was shown that a particularly intuitive analysis of a spin qubit placed in a slowly fluctuating classical magnetic field could be performed by expanding the timedependent field as a Taylor series. $\frac{8}{3}$  While this may seem like a special case, this technique applies to a more general class of pure-dephasing quantum problems in which longitudinal relaxation may be ignored. We investigate the effect of UDD on a spin qubit placed in such a field and show that it is the *n*th UDD sequence that suppresses the effect of all terms up to and including order *n* in the Taylor expansion of the field. These results are applied to the NV center and used to obtain improved sensitivities for NV-based magnetometry.

An NV center interacting with an external magnetic field is described by the Hamiltonian  $\mathcal{H} = \mathcal{H}_{zfs} + \mathcal{H}_{ext} + \mathcal{H}_{int}$ . The first term describes the zero-field splitting of the ground-state Zeeman levels,  $\mathcal{H}_{zfs} = \hbar D S_z^2$ , where  $D = 2.88$  GHz. The interaction with an external magnetic field  $\mathbf{B}_{ext}(t)$  is described by  $\mathcal{H}_{ext}$ . The fields we consider here are small relative to *D* and are hence unable to induce a spin flip, permitting us to ignore all  $S_{x,y}$  terms, giving  $\mathcal{H}_{ext} \approx \gamma B_{ext}^{(z)} S_z$ , where  $\gamma$  is the NV gyromagnetic ratio. For simplicity, we put  $B_{ext}^{(z)} \equiv B_{ext}$ . The final term describes the interaction with the paramagnetic environment of the diamond crystal. As we will see, for resolving the nonunitary dynamics of the reduced density matrix of the NV center, these interactions may be subsumed into a single "internal magnetic field,"  $B<sub>int</sub>(t)$ . We define the fluctuation regime of the external/internal environment via the dimensionless numbers,  $\Theta_{ext} = (\gamma \sigma_0^{ext} \tau_{ext})^{-1}$  and  $\Theta_{int} = (\gamma \sigma_0^{int} \tau_{int})^{-1}$ , where  $\sigma_0^{\text{ext}}/\sigma_0^{\text{int}}$  are the rms field strengths and  $\tau_{\text{ext}}/\tau_{\text{int}}$  are the correlation times of the internal and external environments, respectively. Rapidly and slowly fluctuating fields satisfy  $\Theta$  $\geq 1$  and  $\Theta \ll 1$ , respectively.

## **II. DEPHASING IN THE PRESENCE OF UHRIG DYNAMIC DECOUPLING**

An arbitrary time-dependent magnetic field may be decomposed as a Taylor series in  $t : B(t) = \sum_{k=0}^{\infty} a_k t^k$ . The validity of this expansion rests upon the condition that  $a_{k+1}t^{k+1}$  $a_k t^k$  for all *k*. This is satisfied for  $t < \tau_{int}/\sqrt{2}$ .<sup>8</sup> For times *t*  $\gg \tau_{\text{int}}$  the qubit will exhibit motional-narrowing behavior. In many cases of practical interest,  $B<sub>int</sub>$  is the sum of fields from a large number of dipoles. This implies that the  $\{a_j\}$  are normally distributed with zero mean at room temperature and variance  $\sigma_j^2 = \langle a_j^2 \rangle$ . This leads to the following dephasing envelope:

<span id="page-1-0"></span>
$$
\mathcal{D}(t) = \prod_{j=0}^{\infty} \exp[-(\Gamma_j t)^{2j+2}],
$$
  
where  $\Gamma_j = \left(\frac{1}{\sqrt{2}} \frac{\gamma \sigma_j}{j+1}\right)^{1/(j+1)}$ . (1)

Since  $\Gamma_0$   $\geq \Gamma_k \forall k \geq 1$ ,  $\Gamma_0$  serves to define the free induction decay time,  $T_2^* = 1/\Gamma_0$ . For a 1.1% <sup>13</sup>C bath, the variance of

the magnetic field is given by  $\sigma_0^2 = \sum_i (B_i^2)$ . For a <sup>13</sup>C density  $n_c$  and gyromagnetic ratio  $\gamma_c$ ,  $\sigma_0 \approx \sqrt{\frac{2\pi}{3}} \frac{\mu_0}{4\pi} n_c \hbar \gamma_c \approx 2$   $\mu$ T, and  $T_2^* \approx 4$   $\mu$ s, in good agreement with Refs. [3](#page-4-4) and [15.](#page-4-14) Similarly, using an 0.3% <sup>13</sup>C bath yields  $T_2^* \approx 15$   $\mu$ s, in agreement with Ref. [6.](#page-4-6)

The Hahn-echo sequence removes the effect of a static field on the system as each  $\pi$  pulse effectively sends  $B \rightarrow$  $-B$ . For a field described by  $\Sigma_k a_k t^k$ , this will remove the effect of the  $a_0$  term, and modify all other terms as  $a_i \mapsto (1$  $(-2^{-j})a_j$ <sup>[8](#page-4-2)</sup>. For an NV center, the correlation time of the environment is dictated by interactions between  $^{13}$ C nuclei. A straightforward calculation shows  $\tau_{int} \sim \sqrt{\frac{6}{\pi}} \frac{4\pi}{\mu_0} / (n_c \hbar \gamma_c^2)$ <br>=[1](#page-1-0)5 ms. Using this in Eq. (1) for *j*=1 gives  $\Gamma_1$ =2.1 kHz. We identify  $T_2=1/\Gamma_1=400 \mu s$ , in agreement with Refs. [3](#page-4-4) and [15.](#page-4-14) For an  $0.3\%$  <sup>13</sup>C bath we achieve  $T_2=1.5$  ms as seen in Ref. [6.](#page-4-6) We do not define  $\tau_{int}$  via interaction between <sup>13</sup>C nuclei and any background fields,  $B_0$ , since this manifests as decays and revivals on time scales of  $\tau_r \sim 1/\gamma_c B_0$  and does not represent a true loss of information. Such effects may be mitigated by aligning  $B_0$  along the NV axis.

The phase shift of the NV spin,  $\Delta \phi$ , is proportional to the time integral of the applied field. If pulses *m* are applied at the instants  $t_1, t_2, \ldots, t_m$ , the effect of the pulse sequence on the phase shift will be

$$
\Delta \phi = \gamma \left[ \int_0^{t_1} - \int_{t_1}^{t_2} + \dots + (-1)^m \int_{t_m}^{\tau} \left[ B(t) \right] dt. \right] \tag{2}
$$

<span id="page-1-1"></span>The effect of an arbitrary sequence of pulses on the *j*th term in the Taylor expansion is then

$$
a_j \mapsto a_j \frac{\left[\int_0^{t_1} - \int_{t_1}^{t_2} \cdots + (-1)^m \int_{t_m}^{\tau} \right] t^j dt}{\int_0^{\tau} t^j dt}.
$$
 (3)

For a pulse sequence to suppress the effect of a field to order *n*, the instants at which the pulses are applied must be chosen to ensure

$$
\left[\int_{0}^{t_{1}} - \int_{t_{1}}^{t_{2}} + \cdots + (-1)^{m} \int_{t_{m}}^{\tau} \left] t^{j} dt = 0, \right. \tag{4}
$$

not only for  $j=n$  but for all  $j < n$ .

For example, we may wish to modify our  $\frac{7}{2}$ - $\pi$ - $\frac{7}{2}$  pulse sequence in order to remove the effect of the  $a_1t$  term. If we apply pulses at  $t = \frac{7}{4}$  and  $t = \frac{37}{4}$ , we find that the effects of both  $a_0$  and  $a_1$  terms are suppressed. In general, suppression of all terms up to and including order *n* will require at least  $n+1$   $\pi$ pulses. We define  $\tau_{n,k}$  as the time at which the *k*th pulse is applied in the sequence that suppresses all field components up to, and including, order  $n$ . Evaluation of Eq.  $(3)$  $(3)$  $(3)$  implies that determination of the  $n+1$  elements of the set  $P_n$  $=\{\tau_{n,0}, \ldots, \tau_{n,n+1}\}\$  will require the solution of the following set of  $n+1$  algebraic equations for  $\tau_{n,k}$ :

<span id="page-1-2"></span>
$$
a_0: 2\sum_{k=1}^{n+1} (-1)^{k-1} \tau_{n,k} + (-1)^{n+1} \tau = 0,
$$

$$
a_1: 2\sum_{k=1}^{n+1} (-1)^{k-1} \tau_{n,k}^2 + (-1)^{n+1} \tau^2 = 0,
$$
  
\n
$$
\vdots
$$
  
\n
$$
a_n: 2\sum_{k=1}^{n+1} (-1)^{k-1} \tau_{n,k}^{n+1} + (-1)^{n+1} \tau^{n+1} = 0.
$$
 (5)

To this point, there is some freedom in our choice of pulse sequence, as Eq.  $(5)$  $(5)$  $(5)$  is satisfied to  $n=1$  by both CDD and UDD. However, if we solve Eq. ([5](#page-1-2)) to  $n=2$ , we find  $P_2$  $=\{\frac{1}{4}(2-\sqrt{2}), \frac{1}{2}, \frac{1}{4}(2+\sqrt{2})\}$ , which is the third UDD sequence. It is reasonable to conjecture that the *n*th component of the field,  $a_n t^n$ , will be suppressed by the  $(n+1)$ <sup>th</sup> UDD sequence, which we prove below.

For an interrogation time of  $\tau$ , the time of application of the *k*th pulse in the *n*th UDD sequence is given by

$$
\tau_{n,k} = \tau \sin^2\!\left(\frac{\pi k}{2n+2}\right),\tag{6}
$$

where  $1 \leq k \leq n$ .<sup>[10](#page-4-9)</sup> We wish to show that the  $(n+1)$ th UDD sequence suppresses the effect of all terms up to and including the *n*th term in the Taylor expansion of a time-dependent magnetic field; and that the effect of all terms beyond *n* will be reduced.

The phase accumulation of a spin qubit is proportional to the time integral of the magnetic field to which it is exposed. Each  $\pi$  pulse exchanges the basis states of the qubit Hilbert space, which has the same effect on their relative phase as mapping *B* $\rightarrow$ −*B*. Recall from Eq. ([5](#page-1-2)) that the action of the  $(n+1)$ th Uhrig pulse sequence will be to modify each of the *a<sub>j</sub>* via *a<sub>j</sub>*→[2Σ<sup>*n*+1</sup><sub>i</sub>(-1)<sup>*k*-1</sup>sin<sup>2*j*+2</sup>( $\frac{\pi k}{2n+4}$ )+(-1)<sup>*n*+1</sup>]*a<sub>j</sub>*. To prove our claim, we must first show that

$$
2\sum_{k=1}^{n+1}(-1)^{k-1}\sin^{2m}\left(\frac{\pi k}{2n+4}\right) = (-1)^n.
$$
 (7)

<span id="page-2-1"></span><span id="page-2-0"></span>Using  $N = n + 2$ ,  $\sin(x) = \frac{1}{2i}(e^{ix} - e^{-ix})$ , and expanding as a binomial series in *j*, we arrive at

LHS = 
$$
2^{1-2m} \sum_{j=0}^{2m} (-1)^{j+m} {2m \choose j} \sum_{k=0}^{n-1} e^{i\pi a_{jm}N^k}
$$
, (8)

where  $a_{j m N} = \frac{m-j-N}{N}$ . Note that we have added the  $k=0$  term since  $\sum_{j=0}^{2m} {2m \choose j} (-1)^j = 0$ . Since we have restricted ourselves to  $m \le N-1$ , we have that  $e^{i\pi a_{j m N}} \ne 1$ , so we are free to sum over  $k$  as a geometric series in Eq.  $(8)$  $(8)$  $(8)$ , which is not possible for *m* ≥*N*. This gives LHS= $2^{-2m}(-1)^{2m+N} \sum_{j=0}^{2m} {2m \choose j}$ , which is just the sum of terms in the  $(2m+1)$ th row of Pascal's triangle, and evaluates to  $2^{2m}$ , proving Eq. ([7](#page-2-1)).

Replacing *N* with  $n+2$ , we can see that  $a_i \mapsto 0$ ,  $\forall j \leq n$ . Hence all terms in the Taylor expansion of  $B(t)$  up to and including order *n* are zero. Furthermore, since  $0 \leq \sin^2(x)$  $\leq$  1, the term in the square brackets is always less than 1, hence the effect of the remaining  $a_i$  is reduced. Two immediate consequences are that the  $(n+1)$ <sup>th</sup> UDD sequence will suppress dephasing to *n*th order; and that dephasing effects beyond *n*th order will be reduced. If  $a_0, \ldots, a_n = 0$ , then

<span id="page-2-2"></span>

FIG. 2. (Color online) (a) Dephasing envelopes,  $\mathcal{D}^{(n)}$ , for an NV center in a 1.1% <sup>13</sup>C bath for *n*  $\pi$  pulses. As  $n \to \infty$ ,  $\mathcal{D}^{(n)}$  approaches the Heaviside step function,  $H(\tau_{ext}-t)$ . (b) Effect of the number of  $\pi$  pulses on NV coherence times for different <sup>13</sup>C concentrations. In each case, the coherence time is limited by  $\tau_{\text{int}}$ .

 $\langle a_0^2 \rangle, \ldots, \langle a_n^2 \rangle = 0$ . By Eq. ([1](#page-1-0)) all rates,  $\Gamma_0, \ldots, \Gamma_n$ , are zero.

From the above analysis, we see that the application of a UDD sequence of any order will decrease the intrinsic NV dephasing rate. This allows us to extend the interrogation time, thus improving the sensitivity to an external magnetic field,  $B_{ext}(t)$ . Clearly the dynamics of  $B_{ext}$  will be an important factor in ensuring that the effect of the external field is not also suppressed by the pulse sequence. Simple examples include telegraph signals switching in sync with the UDD sequence, or an ac field of controllable frequency whose nodes coincide with each  $\pi$  pulse, which could be realized by a single spin or ensemble of spins being driven by a controllable microwave field. FC sensitivities to rapidly fluctuating fields will also be improved since fields with correlation times shorter than the interrogation time will not be refocused by the UDD sequence.

## **III. SENSITIVITY ANALYSIS**

We denote the dephasing envelope in the presence of the  $(n+1)$ th pulse sequence as  $\mathcal{D}^{(n)}(t) = \prod_{k=n}^{\infty} \mathcal{D}_k^{(n)}(t)$  [Fig. [2](#page-2-2)(a)]. In the presence of background dephasing described by Eq.  $(1)$  $(1)$  $(1)$ , the minimum induced phase from  $B_{ext}(t)$  that may be measured is  $\Delta \phi(b) = [C\sqrt{N}D^{(n)}]$  $(\tau)^{-1}$ , where *C* describes photon shot noise and imperfect collection<sup>3</sup> and *N* is the number of measurements taken. Typically  $C < 0.3$ , however vast improvements have recently been demonstrated by entangling the NV spin with proximate nuclear spins, permitting repetitive readout of the NV spin state[.16,](#page-4-15)[17](#page-4-16) We now discuss the relevant detection protocols and sensitivities for different fields to which these techniques apply.

#### **A. Sensitivity limits: Controlled telegraph signals**

For a telegraph signal switching between  $\pm B_0$  in sync with each  $\pi$  pulse, the qubit will acquire the maximum possible phase for a given interrogation time,  $\Delta \phi = \gamma B_0 \tau$ . This gives a magnetic field sensitivity of  $\eta_{\rm ss}^{(n)} = B_0 \sqrt{T}$  $= [C \gamma \sqrt{\tau} \mathcal{D}^{(n)}]$  $(\tau)$ <sup>-1</sup>. For all cases where  $\Theta_{\text{int}} \le 1$ , we have that  $\Gamma_k \geq \Gamma_{k+1}$  and  $\Gamma_k > \Gamma_k^{(n)}$  so we may approximate the total dephasing envelope,  $\mathcal{D}^{(n)}$ , by the its leading order contribution. That is,  $\mathcal{D}^{(n)}(t) \sim \exp[-(\Gamma_{n+1}t)^{2n+4}],$  implying the optimal interrogation time is  $\tau = \Gamma_{n+1}^{-1} (4n+8)^{1/(2n+4)}$ . We then find the sensitivity to be bounded above by

<span id="page-3-0"></span>

FIG. 3. (Color online) (a) Fundamental shot-noise sensitivity limits,  $\eta_{\text{ts}}$ , assuming *C*=1 for different <sup>13</sup>C concentrations. Smooth curves show the analytic bound on  $\eta_{ts}$ . Dashed lines show the sensitivity to ac fields. (b) Effect of 50 ns pulse widths. The optimal sensitivity occurs for  $n=13$   $\pi$  pulses.

$$
\eta_{+}^{(n)} = \frac{1}{C\gamma} \sqrt{f_e \Theta_{\text{int}}^{-1/n}}.\tag{9}
$$

This upper bound, together with the actual sensitivity (see below), is plotted in Fig.  $3(a)$  $3(a)$  for an NV. Notice that, as  $\Theta_{\text{int}} \rightarrow 1$ , there is little to be gained by applying UDD.

We now compute the sensitivity of the probe by taking into account the effect of the *n*th order pulse sequence on the *n*th order Taylor coefficient. This results in improved sensitivity beyond that indicated above, as a reduction in the *ak* leads to a reduction in the total decoherence rate, and hence an extended interrogation time.

The dephasing rates are found via Eq.  $(1)$  $(1)$  $(1)$ , where  $\sigma_j^{(n)} \mapsto |2\Sigma_{k=1}^{n+1}(-1)^{k-1}\sin^{2j+2}(\frac{\pi k}{2n+4}) + (-1)^{n+1}|\sigma_j$ . The actual dephasing time due to the combined effect of all the  $\Gamma_k^{(n)}$  will be given by the solution to  $\sum_{k=n+1}^{\infty} (\Gamma_k^{(n)} t)^{2k+2} - 1 = 0$  for *t* and are plotted against the number of pulses used in the sequence in Fig.  $2(b)$  $2(b)$ . From this we see that dephasing times asymptote to the correlation time of the bath. This is to be expected since information lost to the bath cannot be recovered by control of the NV spin alone. Retaining all terms, the sensitivity is then

$$
\eta^{(n)}(\tau) = \frac{1}{\gamma \sqrt{\tau}} \exp\left[\sum_{k=n+1}^{\infty} (\Gamma_k^{(n)} \tau)^{2k+2}\right].
$$
 (10)

By minimizing this expression with respect to  $\tau$  we determine the optimal sensitivity,  $\eta_{ts}^{(n)}$ , as shown in Fig. [3](#page-3-0)(a).

#### **B. ac fields**

In many proposals $2,3$  $2,3$  magnetic resonance techniques are used to drive the sample magnetization at some controlled frequency. For a general sinusoidal field, given by  $B_{ac}$  sin( $vt + \chi$ ), the corresponding sensitivity with which  $B_{ac}$ may be measured is

$$
\eta_{\text{ac}}^{(n)} = \eta_{\text{ts}}^{(n)} \frac{\pi}{2} \left[ \sum_{k=0}^{n} (-1)^{k} \int_{\tau_{n,k}}^{\tau_{n,k+1}} \sin(\nu_n t + \chi_n) dt \right]^{-1}, \quad (11)
$$

where  $\nu_n$  and  $\chi_n$  are the frequency and ac phase offset that minimize  $\eta_{ac}^{(n)}$ . For example, for *n*=1, the NV spin will acquire maximum phase when  $\nu = 2\pi/\tau$  and  $\chi = 0$ . For  $n = 5$ ,  $\nu_5 = 9\pi/2\tau$  and  $\chi_5 = 3\pi/4$ . The optimal ac sensitivity is plotted as a function of the number of pulses in Fig.  $3(a)$  $3(a)$ .

Alternatively, by controlling the power of a proximate microwave field source, we may synchronize environment NMR/electron spin resonance (ESR) control frequencies with the chosen pulse sequence. This allows a piecewise continuous sinusoidal signal to be produced whose nodes coincide with the time of application of each pi pulse [Fig.  $1(c)$  $1(c)$ ], giving a sensitivity of

$$
\eta_{\rm cac}^{(n)} = \frac{\pi}{2} \eta_{\rm ts}^{(n)}.
$$
\n(12)

#### **C. Randomly fluctuating (FC) fields**

Many typical biological samples have a high nuclear spin density, which can result in significant additional dephasing. If the dynamics are fast  $(\Theta_{ext} \ge 1)$ , as in the case of Brownian motion, for example, the additional dephasing may be detected as a perturbation in the dephasing rate. $8$  The dephasing envelope will be modified by a factor of  $\mathcal{D}_{ext}(\tau) =$  $\exp(-\Gamma_{\text{ext}}\tau)$ , where  $\Gamma_{\text{ext}} = \frac{1}{2}\gamma_p^2 \sigma_{\text{ext}}^2 \tau_{\text{ext}}$ . The sensitivity with which  $\sigma_{\text{ext}} = \sqrt{\langle B_{\text{ext}}^2 \rangle - \langle B_{\text{ext}} \rangle^2}$  may be measured is then<sup>8</sup>

$$
\eta_{\rm FC} = 2\Theta_{\rm ext} \eta_{\rm ts}^{(n)},\tag{13}
$$

making the field more difficult to detect as the fluctuation rate increases. This is consistent with motional narrowing phenomena in NMR in which high frequency noise is known to have a reduced effect on the sample  $T_2$  as compared with quasistatic noise. If the dynamics are slow  $(\Theta_{ext} \le 1)$ , the dephasing will be suppressed, permitting the application of the ac methods outlined above.

### **D. Effects of finite width pulses**

Coherent manipulation of the NV is achieved via a resonant ESR transition or Rabi cycle. Instantaneous  $\pi$  pulses cannot be achieved in practice and lead to additional decoherence effects. For a Rabi frequency of  $\Omega$ , the decoherence envelope is given by  $\mathcal{D}_R = [1 + (\gamma^2 \sigma_0^2 t/\Omega)^2]^{-1/4}$ ,<sup>[18](#page-4-17)</sup> and typical pulse errors are  $\approx 1\%$ .<sup>3</sup> Hence for *n*  $\pi$  pulses, the sensitivity will be worsened by a factor of  $\sim 0.99^{-n-1}$ [1  $+\frac{n+1}{4}(\sqrt{\pi}\gamma\sigma_0/\Omega)^4$ , as shown in Fig. [3](#page-3-0)(b). For a pulse width of 50 ns,  $13\pi$  pulses is found to be optimal, with  $\eta_{\rm{ts}}^{(13)}$  $≈ 5.5$  pT Hz<sup>-1/2</sup>.

Cases where the total pulse time is significant compared with the total interrogation time have been considered,<sup>19</sup> however, since we are dealing with extremely long coherence times and short pulse times, these effects have been neglected in this analysis.

### **IV. CONCLUSIONS**

We have theoretically investigated the improvements associated with the application of the optimal UDD sequence to an NV-based magnetometer. Results show that dephasing times are ultimately limited by the self-correlation time of the fluctuating environment, thus NV magnetometer interrogation times may be extended by nearly four orders of magnitude beyond the free-induction decay time. In light of these results, we have shown that incorporation of UDD into current single NV magnetometer protocols may yield sensitivities below 5 pT  $Hz^{-1/2}$  at room temperature in the near future. Such techniques have the potential to yield great improvements to nanoscale sensing, particularly nanobiological processes occurring at room temperature.

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