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We experimentally demonstrate over 2 orders of magnitude increase in the room-temperature coherence time of nitrogen-vacancy centers in diamond by implementing decoupling techniques. We show that equal pulse spacing decoupling performs just as well as nonperiodic Uhrig decoupling and also allows us to take advantage of revivals in the echo to explore the longest coherence times. At short times, we can extend the coherence of particular quantum states out from $T_2^* = 2.7\ \mu$s out to an effective $T_2 > 340\ \mu$s. For preserving arbitrary states we show the experimental importance of using pulse sequences that compensate the imperfections of individual pulses for all input states through judicious choice of the phase of the pulses. We use these compensated sequences to enhance the echo revivals and show a coherence time of over 1.6 ms in ultrapure natural abundance ^{13}C diamond.

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The nitrogen-vacancy (NV$^-$) center in diamond is a model quantum system with coherence times of milliseconds, nanosecond gate times, and an optical handle to allow initialization and readout of single centers [1]. The extraordinary coherence time is essential to proposals for using NV centers in quantum information processing (QIP) [2] or magnetometry [3,4]. However, the long coherence time of the NV defect is not immediately exploitable; one must decouple the electron spin from unwanted interactions with its spin-based environment, that would otherwise lead to a few μs decay time. Decoupling techniques—applying periodic control pulses to suppress the interactions—provide a well understood solution leveraging decades of use in magnetic resonance.

The dominant dephasing mechanism of the NV center in high-purity diamond is the surrounding spin bath of ^{13}C nuclei in the crystal lattice [5]. The NV is an effective spin 1, but a magnetic field lifts the $m_s = \pm 1$ degeneracy and we can concentrate on the effective two-level qubit system of the $m_s = 0/+1$ states. With the magnetic field along the NV symmetry axis, a secular approximation for the effective two-level NV Hamiltonian is given by

$$\mathcal{H}_{\mathrm{NV}} = \omega_S S_z + \sum_j \omega_j I_z^j + S_z \sum_j A_j \cdot \vec{I}^j + \mathcal{H}_{\mathrm{dip.}}, \quad (1)$$

where S/I^j are electron/nuclear spin operators; A_j, the hyperfine coupling to the j'th nuclei; and $\mathcal{H}_{\mathrm{dip.}}$, the dipolar coupling between nuclei. An electron spin superposition state is dephased by time variations in the S_z operator from both fluctuations in the external field and entanglement with the nuclear bath. There is an additional incoherent decay at room temperature when we average over nuclear configurations of the initial maximally mixed state during the $\sim 10^6$ experiment repetitions.

Because these fluctuations are relatively slow, a spin echo (a π pulse with equal delays, τ, before and after) can reverse some of the evolution. However, a key part of the dynamics is that the quantization axis of a ^{13}C spin depends on the state of the electron, due to the anisotropic form of the hyperfine interaction [6]; this gives collapses and revivals in the electron coherence [7]. Because there are no S_x and S_y terms in the secular Hamiltonian, for a free-evolution time τ, the unitary propagator for the electron-bath system is $U_{e,b} = |0\rangle\langle 0|_e \otimes U_{b,0} + |1\rangle\langle 1|_e \otimes U_{b,1}$, with the nuclear bath propagators $U_{b,0/1}$ in the $0/1$ electron manifolds. For the spin-echo sequence with $\rho_{\mathrm{nuc.}} = \mathbb{1}$, the expectation value of the electron superposition state $\sigma_+ = |+\rangle\langle +|$ decays as $\langle \sigma_+ \rangle = \frac{1}{2} + \frac{1}{2}\mathrm{Re}\{\mathrm{Tr}((U_{b,0}U_{b,1})^\dagger (U_{b,1}U_{b,0}))\}$ [5]. If the hyperfine quantization axes are colinear, then $U_{b,0}$ and $U_{b,1}$ commute, and there is an echo for all pulse spacings. However, when the axes are not colinear, then there are additional echo modulations. The initial echo decays on a time scale of a few $\mu\mathrm{s} \approx 1/\sqrt{\sum_j A_j^2}$. We can suppress this decay by rapidly switching between $U_{b,0}$ and $U_{b,1}$ (with electron π pulses) so that they effectively commute—as in a Trotter expansion. At longer times, there are unique circumstances where there is an echo revival. In the $|0\rangle\langle 0|_e$ subspace, where there is no hyperfine interaction, $U_{b,0}$ is dominated by the Zeeman term; hence, it is the same for all ^{13}C nuclei and $U_{b,0} = \exp(-i\tau(\sum_j \omega_j I_z^j + \mathcal{H}_{\mathrm{dip.}})) \approx \mathbb{1}$ at $\tau = n\Omega_L$, integer multiples of the Larmor period, Ω_L. Since $U_{b,0}$ will then factor out of the trace expression, there are echo revivals limited by $\mathcal{H}_{\mathrm{dip.}}$ and any off-axis static magnetic field [5,7,8] [Fig. 4(a)]. We can suppress this decay too by adding more pulses; however, the minimum pulse spacing must be a full Larmor period and the pulse spacings must be commensurate to achieve an echo.

Multiple periodic control pulses have been used to suppress fluctuating fields since the beginnings of magnetic resonance [9]. Its experimental usefulness has been demonstrated in the context of QIP for nuclear bath hyperfine noise for electron spins in the solid state [10–12] and in ion traps [13]. Similar results to those presented here have also been recently reported for the NV system: showing both the importance of compensated sequences to preserve an arbitrary quantum state [14] and the extension of the echo revivals at longer times [15] with the CPMG sequence. Of late, there has been interest in potential improvements offered by nonperiodic spacing of the decoupling pulses. Uhrig dynamical decoupling (UDD) [16], a variant of gradient moment nulling in NMR [17], is provably optimal [18] in certain circumstances. Specifically, UDD is optimal in a time-expansion of the unitary propagator when the noise has a sharp high-frequency cutoff. When the cutoff is relatively softer, conventional CP spacing is preferred [19]. Intriguingly, while suppressing unwanted fluctuations, both approaches can be used to improve the sensitivity of NV magnetometers [4,20].

The noise model for a central spin with varying anisotropic hyperfine couplings is not conventionally studied, except for its pernicious effects on control fidelity [21]. We show experimentally that for this model, equally spaced CP style sequences perform no worse than the optimized pulse spacings of UDD. These experimental tests provide relevant results for the realistic case where there are losses due to pulse imperfections. However, this loss of fidelity from pulse errors is dramatically state dependent. For example, states along the rotation's axis are relatively insensitive. However, for QIP applications the input state may be unknown; this requires compensated sequences robust to pulse errors for all input states. We investigated three decoupling sequences (Fig. 1): conventional CPMG, which for even numbers of pulses compensates for pulse errors along the direction of the rotation axis; the XY family which are compensated for all three axes of the Bloch sphere [22]; and, finally, the UDD which is uncompensated for pulse errors.

We used a standard purpose-built confocal microscope setup for optical initialization and readout of single NV centers in single crystal diamond. We controlled the effective two-level system ($m_s = 0, 1$) with microwaves modulated by an arbitrary waveform generator to provide arbitrary microwave amplitude and phase (see supplementary material [23]).

When the input coherence is parallel to the pulse rotation axis we can explore the extension in T_2 with the number of pulses and the effect of variable pulse spacing (see Fig. 2 and Table I). However, the UDD and XY sequences still suffer from pulse sequence imperfections

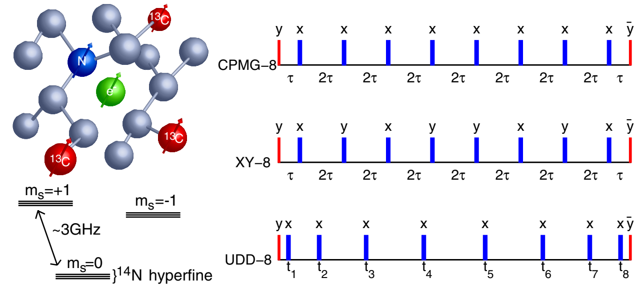

FIG. 1 (color online). NV in a diamond lattice and electron ground state level diagram. The electron spin (green) is coupled to the surrounding nuclear spins: $^{14/15}$N (blue) and ^{13}C (red) in the lattice. An example with eight 180° pulses (in blue) of the three decoupling sequences used: CPMG; XY-8 and UDD. CPMG and XY have the same equal pulse spacings. The timing of the center of the kth UDD pulse with N total pulses is given by $\sin^2(\pi k/(2N + 2))$. Also shown in red are the initial 90° pulse about the y axis, which creates a coherence along the axis of the 180° refocusing pulses, and the final 90° readout pulse.

and there is evidence of an additional early decay with more than 16 pulses.

We posit that the observed performance of the sequences is due to the nature of the non-Markovian spin bath inherent to NV centers in the high-purity diamond relevant for magnetometry and QIP applications. The coherent dynamics of the dilute spin bath is a non-Markovian noise source not easily treatable as a semiclassical fluctuating magnetic field at the electron spin [24]. This is in contrast to recent results in less pure diamond where the dense spin bath and intrinsically shorter T_2's give results that matched the semiclassical theory [14]. We validate our assumption via simulations of the NV center and nuclear spin bath, made tractable through the disjoint cluster method [5]. These simulations provide insight and confirmation of the validity of our experimental results (see supplementary material [23]): that, for example, the extension in coherence we are seeing is not polluted by ensemble or inhomogeneous effects [25,26]. There results help confirm that decoupling techniques, even with real finite length pulses, will continue to be useful in the single to few spin limit with dilute spin bath environments. The simulations confirmed the surprising almost linear increase in T_2 with the number of pulses, n, as opposed to the expected $n^{2/3}$ dependence for a semiclassical field [24] that was recently observed in low-purity diamond [14].

An additional complexity in the NV system is the ^{14}N hyperfine coupling of 2.1 MHz. This leads to substantial coherent off-resonance errors in the π pulses. Our 32 ns, nominally square, pulses have a simulated fidelity of 98.6% (assuming a maximally mixed ^{14}N state). Any experimental implementation will fall short of this due to errors such as fluctuations in the microwave power or transient variations in the microwave phase at the edges of the pulse. For sequences where all the pulses are about the same axis, these errors add coherently and rapidly destroy the

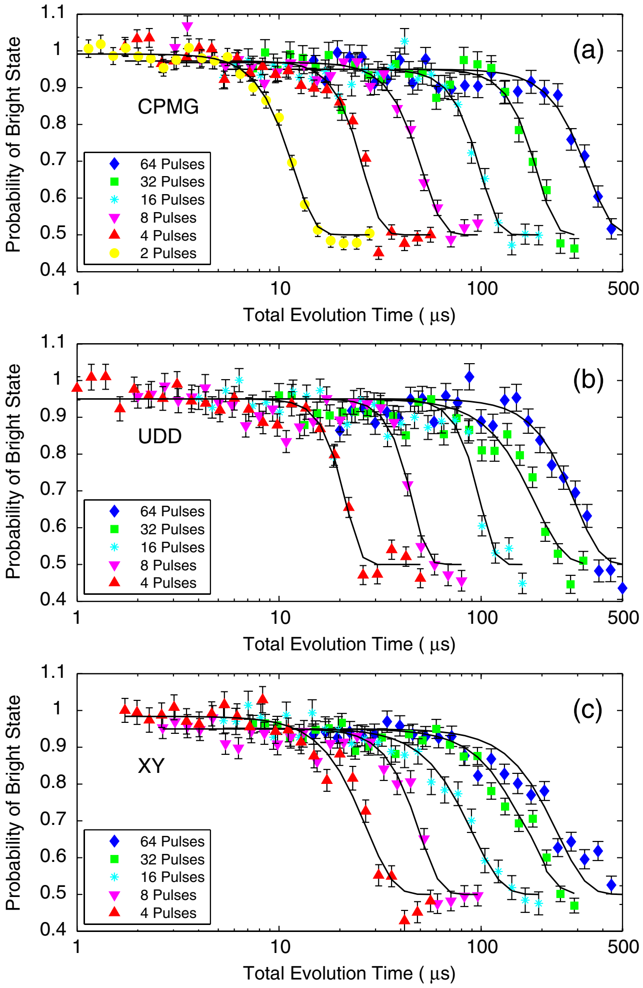

FIG. 2 (color online). Short-time coherence decay for states along the refocusing pulses' rotation axis with (a) CPMG, (b) UDD and (c) XY sequences. Fits are to the phenomenological form $s(t) = 0.5 + A\exp(-(\frac{t}{T_2})^k)$, where $3 < k < 6$. Error bars are propagated from photon counting statistics.

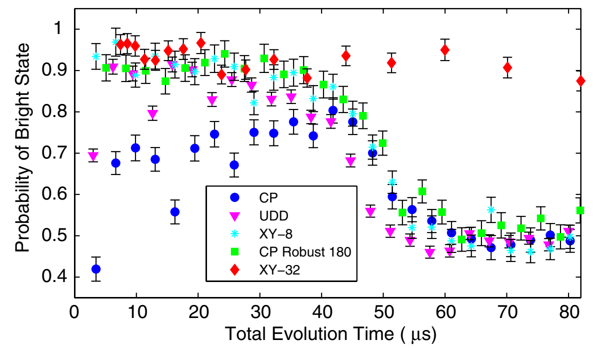

FIG. 3 (color online). CP-8 (circles), and UDD-8 (▼) with the input state perpendicular to the pulses' rotation axis. The deleterious effect of coherent pulse errors leads to initial coherent oscillations that are rapidly damped as they are not properly refocused. Modulating the phases of the pulses as in the XY-8 sequence (★) or using robust composite pulses (■) can overcome this. Also shown is the XY-32 sequence (◇) demonstrating that we can preserve an arbitrary state to much longer times by shortening the pulse spacing.

effectiveness for particular input states. For our setup, beyond 4 pulses, decoupling for states perpendicular to the rotation axis was ineffective (Fig. 3).

TABLE I. Effective T_2 (μs) extracted from the fits to the curves in Fig. 2. For 2 pulses CPMG and UDD are equivalent and there is no identity XY sequence possible. Experiments with a fixed pulse spacing and over 1600 pulses show it is possible to extend this decay out to greater than 800 μs (see supplementary material [23]).

# of pulses	CPMG	UDD	XY
2	11.8 ± 0.4	11.8 ± 0.4	⋯
4	26 ± 2	22 ± 2	27 ± 2
8	51 ± 3	46 ± 3	50 ± 3
16	100 ± 5	99 ± 8	91 ± 6
32	190 ± 13	185 ± 17	168 ± 12
64	340 ± 25	293 ± 25	239 ± 21

One approach is to make the decoupling sequence itself more robust for any initial state: the motivation for the XY family of sequences [22]. By alternating the phase of the pulses about the $\pm X$ and $\pm Y$ axes, and concatenating appropriately, the pulse sequence can be compensated for pulse errors and the overall errors made *isotropic* (Fig. 3). Indeed, our experimental results show that this sequence provides useful refocussing for both input states. The tradeoff for making the errors isotropic is that this sequence will not perform as well as CPMG for a known input state (Fig. 2).

A second approach is to use composite pulse sequences to make the net rotation of individual pulses more robust. We chose the $180_{30} - 180_0 - 180_{90} - 180_0 - 180_{30} = Z_{60}180_0$ sequence (attributed to Dr E. Knill) for its robustness to resonance offsets and ease of calibration. The additional Z rotation is easy to absorb in an abstract reference frame shift. Implementing this composite sequence with 46 ns Gaussian pulses (to avoid overlap of transients at the pulse edges) gives a ideal fidelity of 99.95%, and as seen in Fig. 3, an overall improved performance. Combining composite pulses with XY sequences provides even better performance.

The effect of the multiple pulse echoes can be particularly dramatic when we observe the echo revivals. These revivals will be useful in refocusing the electron spin coherence while performing nuclear gates in QIP applications [2] or magnetometry [15]. The revivals decay due to off-axis fields and nuclear dipole-dipole coupling of up to a few kHz. By applying more than one echo pulse, then we can suppress the decay of the revivals at the expense of less frequent revivals: the revivals only occur when the shortest pulse spacing corresponds to a nuclear identity operation in the $m_s = 0$ manifold. Hence, it is also not possible to see echo revivals with the unequal pulse spacing of UDD. We were able to demonstrate an over sevenfold increase in the

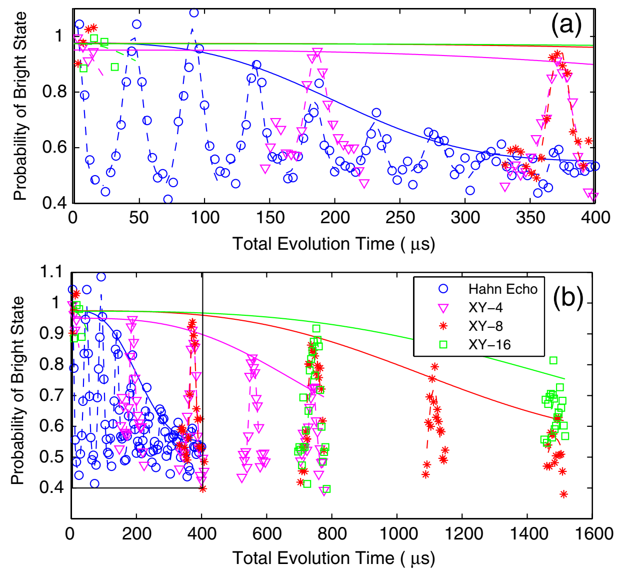

FIG. 4 (color online). XY multiple pulse sequences at echo revivals at short times (a) and longer times (b). The revivals of the single-pulse Hahn echo are observed to decay with a 220 μs T_2. With multiple pulses, the revival frequency is reduced proportionally but the effective T_2 is greatly extended to over 1.6 ms. The envelopes are fit to $s(t) = 0.5 + A\exp(-(\frac{t}{T_2})^3)$. We observe only at revival peaks to collect data faster.

"T_2" over the single-pulse Hahn echo time by using the robust XY-4, XY-8 and XY-16 sequences from 220 μs to over 1.6 ms (Fig. 4). This newly revealed extraordinary coherence time is then comparable to the 1.8 ms reported for isotopically purified diamond, some of the longest room-temperature coherence times for a solid-state electron spin [27].

In summary, we have experimentally demonstrated dramatic increases in the effective dephasing time of a NV center in diamond by using robust sequences and composite pulses to suppress both the fluctuations that lead to the dephasing and the intrinsic errors in the pulses themselves. We expect these robust sequences will prove useful not only in both NV magnetometry and QIP applications but also in other solid-state QIP implementations such as quantum dots [10] or superconducting qubits [28]. The single spin-echo coherence time of our diamond was not particularly long at 220 μs, and as recently reported for the CPMG sequence, even longer coherence times are possible in better diamonds [15] which should help approach the ultimate T_1 limit.

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