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Real-Time Charge Initialization of Diamond Nitrogen-Vacancy Centers for Enhanced Spin Readout

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A common impediment to qubit performance is imperfect state initialization. In the case of the diamond nitrogen-vacancy (NV) center, the initialization fidelity is limited by fluctuations in the defect's charge state during optical pumping. Here, we use real-time control to deterministically initialize the NV center's charge state at room temperature. We demonstrate a maximum charge initialization fidelity of 99.4±0.1% and present a quantitative model of the initialization process that allows for systems-level optimization of the spin-readout signal-to-noise ratio. Even accounting for the overhead associated with the initialization sequence, increasing the charge initialization fidelity from the steady-state value of 75% near to unity allows for a factor-of-two speedup in experiments while maintaining the same signal-to-noise-ratio. In combination with high-fidelity readout based on spin-to-charge conversion, real-time initialization enables a factor-of-20 speedup over traditional methods, resulting in an estimated ac magnetic sensitivity of $1.3\,\mathrm{nT/Hz^{1/2}}$ for our single NV-center spin. The real-time control method is immediately beneficial for quantum sensing applications with NV centers as well as probing charge-dependent physics, and it will facilitate protocols for quantum feedback control over multi-qubit systems.

I. INTRODUCTION

The accelerating pace of quantum technology is evident in the advancement of quantum sensors [1] and the emergence of quantum networks [2]. Critical to these developments have been solid-state spin qubits based on semiconductor defects, due to their optical interface [3], compatibility with integrated technologies [4], and wide selection of host materials [5]. The most well-known example is the nitrogen-vacancy (NV) center in diamond [6, 7], which has enabled pivotal advances in quantum sensing [8–14] and quantum information processing [15–17].

One limitation to the performance of NV-center qubits is imperfect initialization into the oft-desired negative charge state (NV^-). Optical pumping with 532 nm light produces a steady-state statistical charge distribution; typically the probability to prepare the NV^- state is around 75% [18, 19], although it can be much lower for defects close to surfaces [20]. This probabilistic steady-state initialization (SSI) hampers spin readout by decreasing contrast and increasing readout noise [7], and it limits the fidelity of quantum gate operations of coupled spin systems utilizing the NV center as an ancilla [18, 21]. Existing techniques to improve the charge initialization fidelity include doping electrically [22] or chemically [23], and multi-color optical pumping [24]. In addition, many experiments utilize post selection to filter out the noise [20, 24–28]. These techniques either impose strict constraints on materials and device design or require elongated experimental runtime. At cryogenic temperatures, deterministic initialization protocols based on real-time feedback have been essential for entanglement generation and quantum error correction using NV centers due to their long measurement times [16, 29, 30], however these techniques have not been adapted for quantum sensing applications where the duration of each measurement cycle drastically affects the overall sensitivity.

Here, we use real-time feedback to control an NV center's charge-state initialization fidelity at room temperature, and we demonstrate improved spin readout efficiency and sensitivity. A model for the stochastic initialization procedure allows for the selection of near-unity initialization fidelity into either charge state, or an arbitrary intermediate charge distribution. We measure the influence of charge fidelity on the spin readout signal-to-noise ratio (SNR) for two readout techniques, traditional photoluminescence (PL) and spin-to-charge conversion (SCC). Our comprehensive model allows for the optimization of initialization and readout parameters for quantum control experiments of arbitrary durations. The real-time initialization (RTI) protocol improves the spin readout efficiency and reduces the time required for experiments; in combination with SCC readout, we demonstrate a factor-of-20 speedup as compared to traditional methods.

II. EXPERIMENTAL METHODS

A schematic of the experiment is shown in Fig. 1(a). The traditional portion of the setup consists of the lasers, microwave sources, diamond device, and photon-counting

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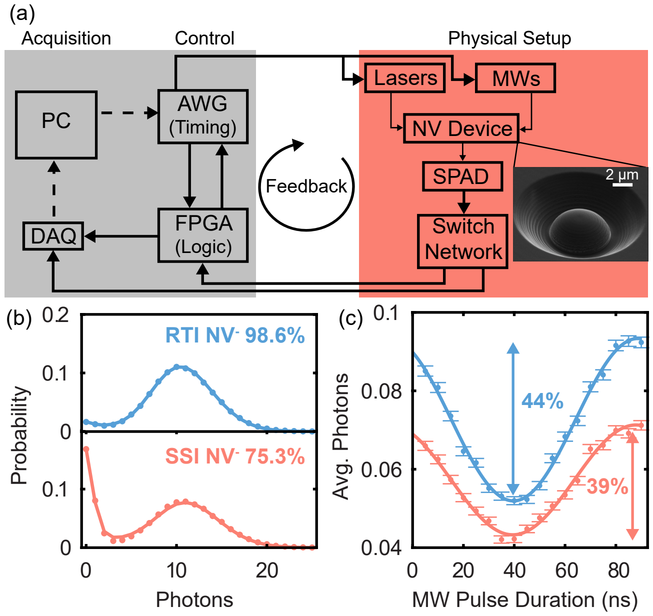

FIG. 1. Real-time charge initialization. (a) System overview for implementing real-time feedback on a nitrogen-vacancy (NV) center's charge state. Inset: scanning electron micrograph of a solid immersion lens fabricated around a single NV center. (DAQ: data acquisition, AWG: arbitrary waveform generator, FPGA: field programmable gate array, MWs: microwaves, SPAD: single-photon avalanche diode). (b) Charge-readout distributions demonstrating the difference in charge state initialization fidelity for the real-time (RTI, top panel) and steady state (SSI, bottom panel) initialization protocols. (c) Rabi nutations of a single NV center following RTI (top, blue curve and data points) and SSI (bottom, salmon curve and data points) demonstrating the increased signal and spin contrast (signified by the arrows). Curves are fits to a sinusoidal oscillation.

electronics. The sample is an electronic grade, type-IIa, synthetic diamond (Element Six) which has been irradiated with 2 MeV electrons (10^{14} cm^{-2}) and annealed at 800°C for 1 hour in forming gas. A solid immersion lens aligned to a single NV center was fabricated using focused-ion-beam milling to increase the photon collection efficiency [24], resulting in a saturated count rate of 300 kCts/s under 532 nm excitation [31]. Imaging and optical control is performed with a home-built room-temperature scanning confocal microscope with three excitation sources. A continuous-wave 532 nm laser (Gem 532, Laser Quantum), referred to as "green," is gated by an acousto-optic modulator (AOM) in a double-pass configuration; it is used for optical pumping and traditional PL readout. An amplitude modulated 635 nm laser diode (MLD 06-01 638, Cobolt), referred to as "red," is used for charge readout and SCC. A continuous-wave 592 nm laser (VFL-592, MPB Communications, Inc.), referred to as "orange," is gated with an AOM and is used for SCC. A 115 G magnetic field is aligned along the NV axis to distinguish the $m_s = \pm 1$ states. A lithographically-defined loop-antenna surrounding the solid immersion lens is driven by an amplified (ZHL-16W-43-S+, Mini-Circuits), amplitude modulated (ZASWA-2-50DR, Mini-Circuits), continuous-wave signal generator (SG384, Stanford Research Systems), which allows for ground-state spin control.

The NV center's charge state is determined to a high accuracy by utilizing a wavelength that excites the NV$^-$ zero phonon line of 637 nm but not the NV^0 zero phonon line of 575 nm [25]. Example histograms of photon counts arising from 75,000 charge readouts are shown in Fig. 1(b) for both the steady-state NV$^-$ population of $75.3 \pm 0.4\%$ and a higher fidelity initial population of $98.6 \pm 0.2\%$. These populations were determined by fitting to a statistical model describing the observed photon number histogram [28]. The SSI value of $\sim 75\%$ agrees with previous measurements [19]. The benefit of this elevated initialization fidelity can be seen in the ground-state Rabi nutations in Fig. 1(c), where the higher purity charge state exhibits higher brightness and contrast.

We implement real-time control by linking our timing electronics, which consist of an arbitrary waveform generator (AWG, AWG520 Tektronix) and data acquisition (DAQ, National Instruments) system, with the fast digital logic of a field programmable gate array (FPGA, Virtex-7 Xilinx); refer to Figure 1(a) for the full system overview. In the initialization control loop, the AWG outputs a sequence consisting of a green pump and red charge probe in an repeating loop; when the FPGA detects that a preset photon detection threshold has been reached during the charge probe, it sends an event signal to advance the AWG out of its loop and continue with the other predefined measurements. The time it takes from detection of the final photon to the halting of the initialization procedure is $\tau_{\text{delay}} = 550$ ns, which consists of the detector delay (30 ns), the AWG delay (500 ns), and the red laser delay (20 ns).

III. RESULTS

A. Real-time charge initialization

We model the charge probe process using a photon distribution model accounting for transitions between NV$^-$ and the neutral (NV^0) charge state [28, 32, 33]. The model assumes that the charge dynamics of the NV center can be reduced to a two-state system with emission rates γ_- and γ_0, and charge transition rates for ionization (negative to neutral, Γ_{Ion}) and recombination (neutral to negative, Γ_{Rec}); see Fig. 2(a). We determine these rates as a function of power by measuring the photon distributions during a time bin that allows for about one ionization event to occur and fitting to the model [31]. Since the charge readout powers used in this work are below the saturation regime, the emission rates scale linearly with laser power while the ionization and recombination rates scale quadratically with power [18, 19].

The control parameters governing the charge probe

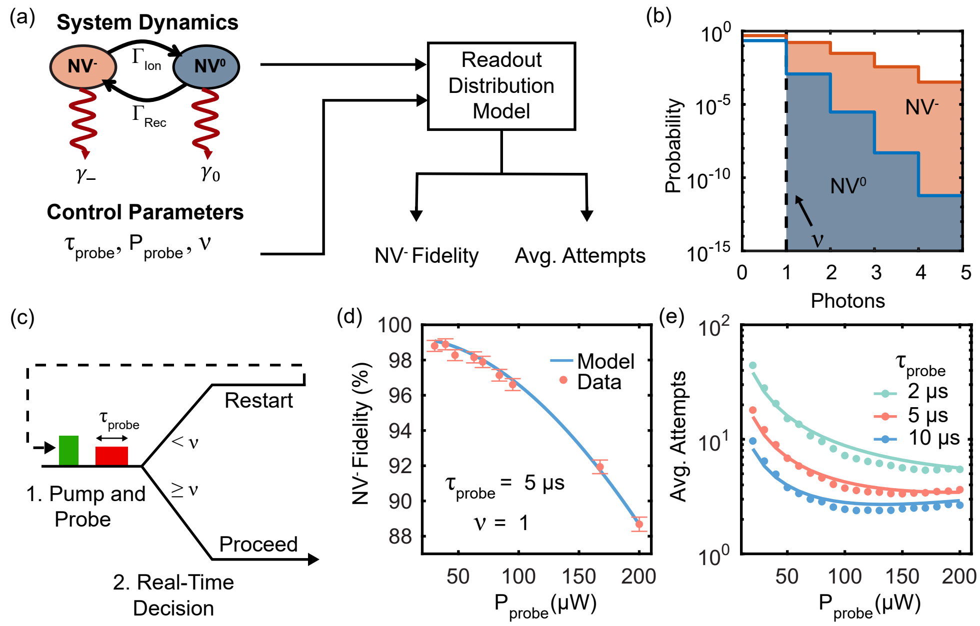

FIG. 2. Modeling real-time control. (a) Conceptual diagram of the model. The system dynamics model describes how the charge-dependent emission rates (γ_-, γ_0) and charge interconversion rates (Γ_{Ion}, Γ_{Rec}) depend on illumination power ($\mathrm{P_{probe}}$). Given a readout duration (τ_{probe}) and a threshold condition ($\nu_{\mathrm{threshold}}$), the readout distribution model determines the NV$^-$ fidelity and the average number of attempts required to reach the threshold. (b) Modeled photon distributions for the two charge states with $\tau_{\mathrm{probe}} = 5\,\mu\mathrm{s}$ and $P_{\mathrm{probe}} = 100\,\mu\mathrm{W}$. (c) Experimental timing diagram and decision tree for initializing the charge state. (d) Comparison between the modeled (line) and measured (markers) NV$^-$ fidelity as a function of probe power. (e) Comparison between the modeled (lines) and measured (markers) average attempts to reach the threshold ($\nu = 1$) as a function of powers and probe duration. Error bars in (e) are comparable to the marker size.

process are the laser power ($\mathrm{P_{probe}}$), maximum duration (τ_{probe}), and the photon threshold (ν) that defines the termination condition of the initialization loop [Fig. 2(a)]. Given these three parameters, the model provides the expected photon distributions for the negative or neutral charge state configurations,

$$p(n|s), \tag{1}$$

where n is the number of photons detected during τ_{probe} and $s = -$ or 0 signifies the initial charge state; see Fig. 2(b) for an example.

The distributions allow us to calculate two critical metrics for RTI: the NV$^-$ charge fidelity (F_{NV^-}) and the average attempts ($\bar{n}$) required for successful initialization. The initialization fidelity is governed by two terms,

$$F_{\mathrm{NV}^-} = (1-\epsilon_{\mathrm{T}})(1-\epsilon_{\mathrm{D}}), \tag{2}$$

where ϵ_{T} is the threshold error and ϵ_{D} is the delay error. The threshold error is the probability that NV^0 leads to a threshold reaching event and is given by

$$\epsilon_{\mathrm{T}} = \frac{\sum_{n\geq\nu}(1-P_-)p(n|0)}{\sum_{n\geq\nu}\left[P_-p(n|-)+(1-P_-)p(n|0)\right]}, \tag{3}$$

where P_- is the probability that the NV center was initially in NV$^-$ prior to the charge probe. The delay error is the probability that an ionization event occurred during the electronic delay time and is given by

$$\epsilon_{\mathrm{D}} = 1 - e^{-\tau_{\mathrm{delay}}\Gamma_{\mathrm{Ion}}}. \tag{4}$$

The average attempts to initialize is given by

$$\bar{n} = \left(\sum_{n\geq\nu} P_-p(n|-)+(1-P_-)p(n|0)\right)^{-1}. \tag{5}$$

As an ensemble average, $\bar{n}$ takes continuous values.

Figure 2(c) outlines the experimental decision tree in the real-time initialization procedure. A charge pump-and-probe sequence is repeatedly played out by the AWG until the FPGA detects a threshold reaching event. The green pump pulse is set to $500\,\mu\mathrm{W}$ and $500\,\mathrm{ns}$ to quickly repump the charge without incurring significant overhead; we vary P_{probe} and τ_{probe} to optimize the performance. In order to verify our model, we measure F_{NV^-} and $\bar{n}$ as a function of P_{probe} as shown in Figs. 2(d) and (e). We extract F_{NV^-} by performing a subsequent charge measurement and fitting to the photon distribution model, and determine $\bar{n}$ from the time it takes to record 10^5 threshold reaching events.

The measurements of F_{NV^-} are generally consistent with our model. We attribute the minor discrepancy between the measured values of $\bar{n}$ and the model predictions

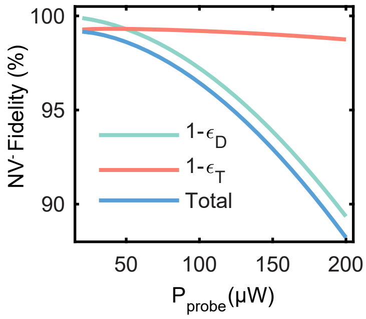

FIG. 3. Initialization errors. Modeled F_{NV^-} as a function of $\mathrm{P_{probe}}$ together with the independent contributions due to thresholding and delay errors. The model parameters are the same as in Fig. 2(d).

to minor variations in the steady-state charge population imposed by the control sequence. The model assumes a fixed initial NV$^-$ population of $P_- = 75\%$, however we observe that the initial population depends weakly on the probe duration and power used in a repeated experiment. We neglect this higher-order effect since it has the beneficial effect of decreasing $\bar{n}$ for the control parameters we employ.

The relative contribution of the two error sources in the charge initialization fidelity depend on $\mathrm{P_{probe}}$; see Fig. 3. At low powers, ϵ_{T} is dominant and F_{NV^-} is limited by the signal-to-background ratio of the charge readout process. For a threshold of 1 photon, the maximum achievable fidelity is $98.6 \pm 0.2\%$ as $\mathrm{P_{probe}}$ approaches zero, however $\bar{n}$ becomes large; see Fig. 2(e). At higher powers, ϵ_{D} is dominant due to the quadratic scaling of the ionization rate with power. Therefore, when designing an experiment utilizing RTI that is sensitive to timing overheads, it is crucial to minimize the control delay time in order to maintain high initial fidelity along with a small $\bar{n}$.

To verify that RTI preserves the ground state spin properties, we measured the coherence times for Ramsey (T_2^*) and Hahn echo (T_2) measurements, as well as the spin relaxation time (T_1) [31]. We observe a $\sim 16\%$ increase in T_2^* when utilizing RTI, which could be due to ionization of nearby neutrally charged substitutional nitrogen donors ($S = 1/2$) to the positive charge state ($S = 0$) [34]. We detect no statistically significant difference in T_2 or T_1.

B. Spin-readout performance

We now consider the effect of the initial F_{NV^-} on the spin readout SNR. Generally, the observable for a spin measurement of an NV center follows the form

$$\langle S_i \rangle = \langle \tilde{S}_i \rangle \, F_{\mathrm{NV}^-} + \langle \epsilon \rangle \, (1 - F_{\mathrm{NV}^-}), \tag{6}$$

where $\langle S_i \rangle$ is ensemble-averaged value of the observable S for the spin state i, $\langle \tilde{S}_i \rangle$ is the expectation value of the observable for spin state i given an initial NV$^-$ state, and $\langle \epsilon \rangle$ is an error in the observable which is due to the NV center residing in NV^0 during the readout. The single-shot SNR for spin readout is then given by

$$\mathrm{SNR} = \frac{|\langle S_0 \rangle - \langle S_1 \rangle|}{\sqrt{\sigma_0^2 + \sigma_1^2}}, \tag{7}$$

where σ_i is the standard deviation associated with $\langle S_i \rangle$ [7].

To make quantitative comparisons between readout techniques, the physical observable and its accompanying statistical model must be incorporated into equation (7). For PL readout [Fig. 4(a)], the signal is the average number of detected photons during the first 250 ns of 532 nm illumination and thus obeys Poissonian statistics. For SCC readout [Fig. 4(b)], the signal is the probability of detecting NV$^-$ following the conversion, and it obeys Binomial statistics.

Figure 4(c) details the measurement timing diagram that allows for the characterization of spin SNR as a function of F_{NV^-}. Following initialization with an arbitrary F_{NV^-}, the spin state is either left in the polarized $m_s = 0$ state, or flipped to the $m_s = -1$ state with a 40 ns microwave π-pulse. We estimate the value of $\langle S_i \rangle$ from repeated measurements using both traditional and SCC readout techniques. We also measure the spin SNR for the traditional SSI consisting of 2 μs of 532 nm illumination. We separately optimize PL and SCC readout parameters to ensure a fair comparison between the techniques [31]. The raw data are fit using equation (6), from which we empirically determine $\langle \tilde{S}_i \rangle$ and $\langle \epsilon \rangle$. Figure 4(d) depicts the results of this measurement for both readout protocols, with the SNR calculated using equation (7) for both the data (symbols) and fits (curves).

Interestingly, the spin SNR following RTI for both SCC and PL readout, when controlling for NV$^-$ fidelity, is $\sim 7\%$ higher than for SSI. This is attributed to improved optical spin polarization in the real-time protocol, since the red laser induces negligible recombination; this is consistent with previous observations [35]. The initial spin purity, estimated from measurements of the excited-state lifetime, is approximately 91% and 94% for the steady state and real-time protocols, respectively [31, 36, 37].

C. Spin readout efficiency

By combining the RTI model with the spin SNR as a function of F_{NV^-}, we can optimize the signal acquisition for a given experiment. To achieve this, we define the spin readout efficiency,

$$\xi = \frac{\mathrm{SNR}}{\sqrt{\tau_{\mathrm{I}} + \tau_{\mathrm{O}} + \tau_{\mathrm{R}}}}, \tag{8}$$

where τ_{I} is the initialization time, τ_{O} is the spin operation time, and τ_{R} is the spin readout time. This figure of merit

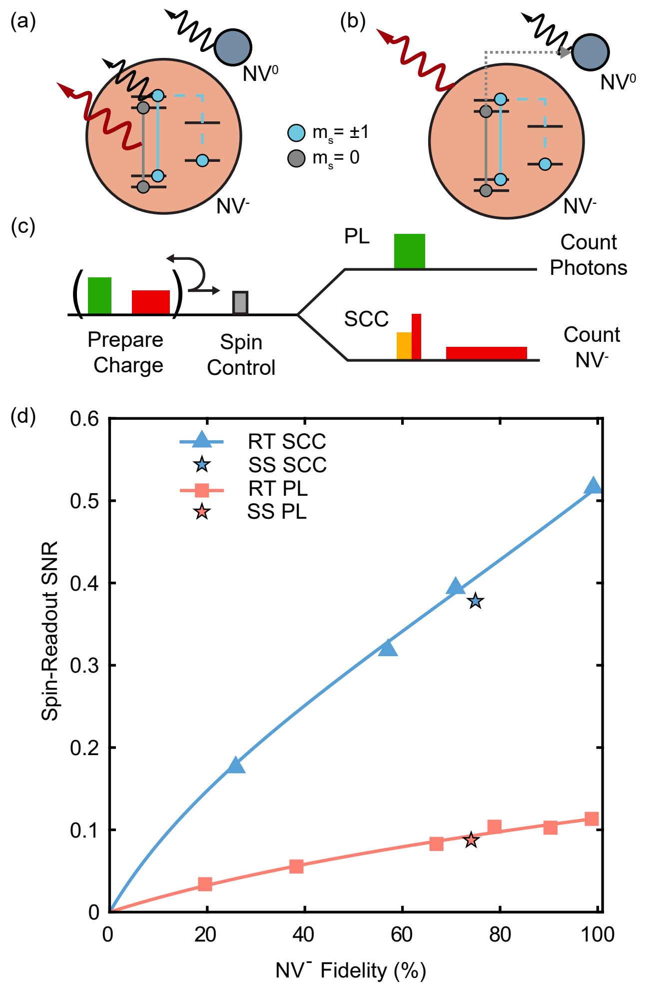

FIG. 4. Spin-readout performance. (a,b) Conceptual diagram for traditional PL readout (a) and SCC (b). Red curves represent the desired signal and black curves contribute to background. (c) Timing diagram for measuring the spin signal-to-noise ratio (SNR) given different heralded charge fidelity. (d) Spin-readout SNR as a function of NV^- fidelity for different initialization and readout techniques. The solid lines represent a fit of equation Eq. 6 to the data, where the fit and data are converted to SNR using Eq. 7. Error bars are comparable in size to the markers.

is related to the sensitivity, and encompasses the single-shot SNR, the spin operation duration, and the associated initialization and readout overheads [7]. The total SNR after multiple measurement cycles with a total integration time, T, is simply given by $\langle \mathrm{SNR} \rangle = \xi\sqrt{T}$. We assume the operation time is fixed by the desired sensing or computation protocol. We have previously considered the optimization of the readout duration, power, and threshold for SCC, and we include those procedures when necessary [7, 38].

Real-time control allows for additional design flexibility in an experiment, as longer time spent initializing results in a higher spin readout SNR yet fewer total averages. Equation (8) quantitatively captures the trade-off between these two quantities. The initialization time is given by

$$\tau_{\mathrm{I}} = (\tau_{\mathrm{pump}} + \tau_{\mathrm{overhead}} + \tau_{\mathrm{probe}})\bar{n}, \tag{9}$$

where $\tau_{\mathrm{pump}} = 0.5\,\mu\mathrm{s}$ is the duration of the 532 nm charge reset pump and $\tau_{\mathrm{overhead}} = 1.5\,\mu\mathrm{s}$ is the overhead in the initialization sequence comprised of the green AOM delay, singlet decay time, and τ_{delay}. Note that τ_{I} is an average quantity since equation (8) is assumed to be an ensemble average over many trials.

With a model describing the readout efficiency, we can numerically optimize equation (8) to determine the protocol parameters that maximize the readout efficiency for a given operation time. To assess the results in context of typical NV-center experiments, we compute and measure the baseline readout efficiency, ξ_{baseline}, corresponding to steady state initialization and traditional PL readout for different operation times. We then define the speedup as the reduction in integration time required to achieve a fixed SNR when comparing a new technique to the baseline,

$$\mathrm{Speedup} = \left(\frac{\xi}{\xi_{\mathrm{baseline}}}\right)^2. \tag{10}$$

A speedup of unity defines the break-even time, τ_{BE}, the operation time at which it is equally efficient to use the enhanced technique over the baseline protocol.

Figure 5 presents the results of this optimization for four different scenarios: SSI with PL readout, RTI with PL readout, SSI with SCC readout, and RTI with SCC readout. The predicted and measured speedup curves for PL and SCC readout are shown in Figs. 5(a) and (b), respectively. For PL readout, we observe a break-even time for using RTI of $\tau_{\mathrm{BE}} \sim 70\,\mu\mathrm{s}$, and a maximum speedup of 1.74 ± 0.09 for an operation time of 1 ms. Interestingly, we find that our full model always results in a choice of measurement parameters that make SCC more efficient than PL readout. RTI offers a further boost for operation times over 30 μs, with a maximum observed speedup of 20.8 ± 1.2 for $\tau_{\mathrm{O}} = 1$ ms. The measurements agree with the model prediction when accounting for the uncertainty in calibrating the single-shot SNR.

Figure 5(c) shows the total SNR as a function of integration bandwidths for each of the four techniques. Here, we have fixed the operation time to be 500 μs. In each case, the total SNR scales with the inverse square root of bandwidth as expected. Of note is the integration bandwidth for which each technique achieves $\langle \mathrm{SNR} \rangle = 1$, which represents the maximum frequency of environmental dynamics that can be resolved above the noise. The RTI protocol coupled with SCC readout offers the best performance for this operation time. In addition, Fig. 5(c) confirms that the optical pulse sequences required for RTI and SCC do not introduce any appreciable noise in the bandwidth we consider.

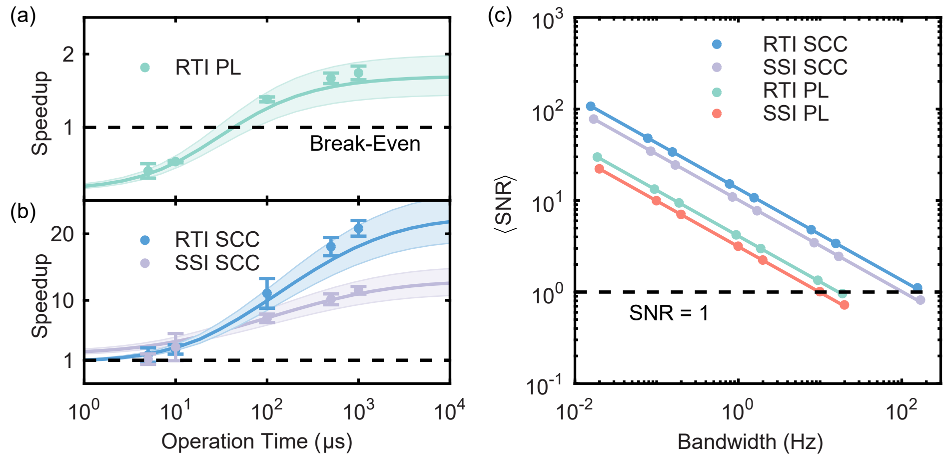

FIG. 5. Performance improvements for time-averaged measurements. Predicted (curves) and measured (data points) speedup for PL spin readout with RTI (a) and SCC readout using SSI and RTI (b). The dashed black line in panels (a) and (b) indicates the break-even condition in comparison to SSI and PL readout. The shaded regions represent 1σ confidence intervals of the model, accounting for uncertainty in the measured single-shot SNR. (c) Total, time-averaged SNR as a function of integration bandwidth for each protocol. Lines are fits to the inverse square root of the bandwidth. Error bars in (c) are smaller than the markers.

IV. DISCUSSION

A. Sensitivity improvements

NV-center quantum sensors stand to gain significant sensitivity improvements from using RTI protocols of the charge state. The largest speedup is realized for long operation times that approach 1 ms, which coincide with the typical requirements for spin relaxometry [14, 39] as well as dynamical decoupling sequences [40, 41].

The single NV center studied here exhibits a Hahn-echo $T_2 \approx 800\,\mu s$ [31]. We estimate the ac magnetic-field sensitivity for our NV center using the expression

$$\eta_{AC} = \frac{\pi\hbar}{2g\mu_B}\sqrt{\frac{T_2+\tau_I+\tau_R}{(T_2)^2}}\sigma_R, \tag{11}$$

where g is the gyromagnetic ratio, μ_B is the Bohr magneton, and σ_R is the spin readout noise [28]. The spin readout noise is directly related to the single-shot SNR through the following expression [7]

$$\sigma_R = \sqrt{1+\frac{2}{\mathrm{SNR}^2}}. \tag{12}$$

Our optimization routine for $\tau_O = T_2 = 800\,\mu s$ yields the following parameters for RTI SCC: $\tau_I = 43\,\mu s$, $P_{\mathrm{Init.}} = 53\,\mu W$, $\tau_R = 127\,\mu s$, $P_{\mathrm{Readout}} = 22\,\mu W$. With these parameters, the predicted SNR $= 0.4$ corresponds to $\sigma_R = 3.67$ and $\eta_{AC} = 1.3\,\mathrm{nT/Hz}^{1/2}$. The optimal parameters from the model correspond to a charge-initialization fidelity of $F_{NV^-} = 98.6\%$, and a charge-readout fidelity of 70%. Interestingly, the charge-readout fidelity is significantly below the achievable maximum of $\approx 95\%$. This implies that, for this operation time, it is advantageous to perform additional averaging of less accurate measurements.

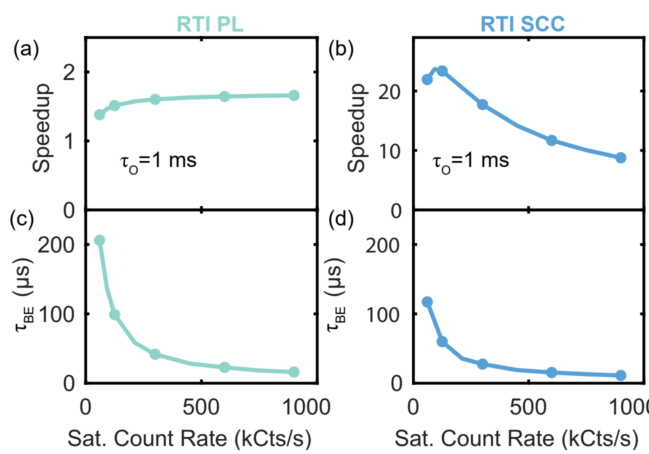

FIG. 6. The effect of photon collection efficiency. (a,b) Predicted speedup for $\tau_O = 1\,\mathrm{ms}$ using RTI together with PL readout (a) or SSC readout (b) as a function of saturated photon count rate under 532 nm illumination. (c,d) Predicted break-even time, at which it becomes advantageous to use RTI, for the same parameters as in (a,b). Points represent the experimentally-relevant count rates mentioned in the text.

We have emphasized the use of RTI together with SCC readout, since the two methods have similar experimental requirements. However, the benefits extend to other NV-center readout techniques used in quantum sensors, such as the nuclear-assisted method [10, 42], which would see similar signal-acquisition improvements due to RTI of the charge state.

B. Gains for NV centers near surfaces

In many situations, the gains from RTI are likely to be even larger than we have demonstrated, since NV centers located in nanodiamonds or close to surfaces typically exhibit lower steady-state charge populations than those in bulk diamond [20, 38]. Using our model and assuming a 25% NV^- steady-state population, RTI would enable a factor-of-6 speedup for PL readout and a factor-of-75 speedup for SCC readout with an operation time of 500 µs.

C. Role of photon collection efficiency

For any quantum device based on PL detection, performance depends critically on photon collection efficiency [7]. The need to maximize photon count rates forces tradeoffs in device design, optical complexity, and versatility of NV centers in different applications. Figure 6 shows the predicted speedup and corresponding break-even times for the use of charge-state RTI, across a range of values for the saturated PL count rate from a single NV center corresponding to typical experimental situations. The predictions in Fig. 6 are calculated by scaling the parameters in our model that depend on photon collection efficiency, corresponding to our measured saturated count rate under 532 nm illumination of 300 kCts/s [31]. Of note are the experimentally-relevant saturated count rates of 60 kCts/s, 125 kCts/s, 600 kCts/s, and 900 kCts/s. Respectively, these values approximate an NV center imaged through a planar diamond surface with a high-NA air objective, a planar sample using an oil-immersion objective [43], a ⟨111⟩-oriented diamond with a solid-immersion lens and air objective [44], and a diamond nanobeam [28]. These values are marked as points in Fig. 6.

Notably, the speedup from RTI persists across this range of relevant photon collection efficiencies. The speedups in Figs. 6(a) and 6(b) are all calculated according to Eq. (10) in comparison to a baseline of SSI PL predicted for the corresponding experimental situation with $\tau_O = 1$ ms. The speedup for PL readout is essentially independent of collection efficiency; for SCC readout the gains are largest for systems with low collection efficiency, where the relative improvement of using SCC over PL readout is magnified. The combined approach of RTI SCC still yields nearly an order of magnitude speedup even for count rates approaching 1 MCts/s.

The break-even times shown in Figs. 6(c) and 6(d) correspond to the value of τ_O for which the spin readout efficiency using RTI equals that for SSI, using the corresponding readout protocol. Increasing collection efficiency results in improved charge initialization fidelity, and a corresponding reduction in τ_{BE}. For high-collection-efficiency devices with saturated count rates approaching 1 MCts/s, the break-even time for using RTI is reduced to 16 µs and 11 µs for PL and SCC, respectively.

D. Maximizing initial state fidelity

While we have focused on applications that require consideration of the overhead from initialization and readout, the RTI technique can be readily adapted to situations in which initialization fidelity is prioritized over total measurement time. For example, the initialization error can be reduced by a factor of 2 by increasing the threshold to 2 photons, and the delay error can be reduced by decreasing P_{probe}. Using $\nu = 2$ and $\tau_I = 7$ ms in our setup, we measure $F_{NV^-} = 99.4 \pm 0.1\%$ [31]. Such control over the charge state could facilitate precise measurements of the local electrostatic environment [20, 45], aid in the quantification of photon collection efficiency for photonic devices [46], and improve the single-shot SNR for infrequent SCC measurements [47]. In addition, the fidelity associated with initializing, controlling, and measuring coupled nuclear spins [17, 21, 30] is intricately tied to the NV center's charge and spin purity and thus could be improved with RTI.

V. CONCLUSION

In conclusion, we demonstrated an efficient method for initializing the charge state of an NV center in real-time and assessed how this can be used to improve the efficiency of spin readout. Real-time control could be applied to other aspects of the NV center, such as projective initialization of nuclear spins [48] and increasing the spin state initialization fidelity through time-gating. In addition, this advanced control can be applied to other emerging solid-state spin defects, especially those which may have a high fidelity readout mechanism but a less-than-ideal spin or charge pumping transition.

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