Quantum optimal control can play a crucial role to realize a set of universal quantum logic gates with error rates below the threshold required for fault-tolerance. Open-loop quantum optimal control relies on accurate modeling of the system under control, and does not scale efficiently with system size. These problems can be avoided in closed-loop quantum optimal control, which utilizes feedback from the system to improve control fidelity. In this paper, two closed-loop quantum optimal control algorithms, the hybrid quantum-classical approach (HQCA) described in [Phys. Rev. Lett. 118, 150503 (2017)] and the finite-difference (FD) method, are experimentally investigated and compared to open-loop quantum optimal control. We employ a solid-state ensemble of coupled electron-nuclear spins serving as a two-qubit system. Our experimental results demonstrate closed-loop quantum optimal control outperforms open-loop quantum optimal control. Furthermore, simulations reveal that HQCA is more robust to measurement noise than the FD method, but is susceptible to control field distortions due to hardware limitations.

In this paper, we experimentally investigated the two gradient-based closed-loop quantum optimal control approaches, HQCA and FD, in a solid-state electron spin resonance (ESR) two-qubit system. The ensemble two-qubit system consists of hyperfine coupled electron and nuclear spins. This system combines advantages of electron spins and nuclear spins, i.e. large thermal polarization and fast control of clear spins, i.e. large thermal polarization and fast control of electron spins, and long coherence time of nuclear spins. It has been shown that universal quantum control using only microwave excitation in this system is possible [19, 21]. However, when designing optimal control pulses for ESR systems, the hardware transfer function cannot usually be ignored. The HQCA method does not explicitly consider the transfer function, so it is important to test its performance experimentally and compare it with the FD method, which in principle accounts for the transfer function. We used two basis sets for the FD method, linear and Slepian [22, 24]. The Slepian basis is designed for limited control bandwidth, and therefore can give further insights regarding the effects of the transfer function bandwidth. Finally, open-loop quantum optimal con-
control was also implemented for comparison. In general, higher fidelities were observed with closed-loop control. Although the HQCA and FD methods showed similar experimental performance, we used simulations to find favorable conditions for each method. When the error is dominated by the transfer function, the FD method performs better. When the error is dominated by the measurement noise, the HQCA method performs better.

Two methods for deriving gradients—The control problem we consider here is to prepare a desired state starting from a given initial quantum state. We choose the state fidelity defined in Eq. (1) as the metric to evaluate the control quality.

\[ F = \frac{\text{Tr}[U(T)\rho f U(T)\dagger\rho f]}{2^n}, \]  
(1)

where \( \rho_i \) and \( \rho_f \) are the initial and target states, respectively, \( T \) is the total duration of the control sequence, and \( n \) is the number of qubits. \( U(t) \) is the unitary evolution of the spin system in the presence of the system’s internal Hamiltonian \( H_0 \) and control Hamiltonian \( H_c(\mathbf{u}(t)) \), and hence satisfies:

\[ \dot{U}(t) = -i[H_0 + H_c(\mathbf{u}(t))]U(t), \]
\[ U(0) = I \otimes I. \]  
(2)

Here \( \mathbf{u}(t) \) is the collection of control parameters, e.g. control field amplitudes, and \( F \) is the 2-dimensional identity operator. The goal is to maximize the fidelity defined in Eq. (1).

Gradient ascent pulse engineering (GRAPE) [25] is a well-known iterative numerical method to solve the optimization problem, where at the \( q \)th iteration the control parameters are updated by

\[ \mathbf{u}^{q+1} = \mathbf{u}^q + c_q \mathbf{g}^q, \]  
(3)

where \( \mathbf{g}^q \) is the gradient of \( F \) with respect to the control parameters \( \mathbf{u}^q \) and \( c_q \) is an adaptive step size. Convergence happens at certain local optima and the solution can be accepted once the desired \( F \) is realized with the parameters \( \mathbf{u}^q \). In this paper, we refer to a class of numerical optimization methods which uses classical resources to calculate the fidelity \( F \) and its gradients \( \mathbf{g}^q \) (i.e. GRAPE) as open-loop quantum optimal control.

One drawback of open-loop quantum optimal control is that it relies on accurate determination of \( H_0 \) and \( H_c \), which can be difficult to obtain in real systems. Moreover, numerical methods become impractical when the size of the system is larger than a handful of qubits [12]. To address such issues, Li et al. proposed a closed-loop quantum optimal control scheme known as HQCA which utilizes the quantum system under control as a quantum simulator in calculating the gradient \( \mathbf{g}^q \). In the following, we briefly describe the method.

The HQCA approach can be applied to many quantum systems. Here, we take the spin-based magnetic resonance as an example. Consider a common control Hamiltonian in magnetic resonance systems, where the control magnetic field is in the transverse plane relative to the static magnetic field, i.e. in the \( x-y \) plane:

\[ H_c(m) = \sum_{i=1}^{N} [u_{x,i}(m)\sigma_x^i + u_{y,i}(m)\sigma_y^i], \]  
(4)

where \( N \) is the number of spins that can be excited by the resonant alternating current (AC) magnetic field, \( \sigma_x^i \) is the Pauli operator of the \( i \)th spin, and \( u_{x,i}(m) \) is the Pauli magnetic field amplitude. Here, we take the spin-based magnetic resonance as an example. Consider a common control Hamiltonian in magnetic resonance systems, where the control magnetic field is in the transverse plane relative to the static magnetic field, i.e. in the \( x-y \) plane:

\[ H_c(m) = \sum_{i=1}^{N} [u_{x,i}(m)\sigma_x^i + u_{y,i}(m)\sigma_y^i], \]  
(4)

Figure 1. (Color online) Flow diagram of gradient-based closed-loop optimal control as applied to ESR. Arrows label the direction of information flow. Error sources are labeled using orange dash-line circles. Both the HQCA and FD methods can compensate for the control errors caused by the uncertainty in the system Hamiltonian. Theoretically, the FD method finds the gradient \( \partial F/\partial \mathbf{u} \), while HQCA finds the gradient \( \partial F/\partial \mathbf{u} \). The pulse shape represented by \( \mathbf{u} \) is distorted by the hardware.

Figure 1 shows the schematic of how closed-loop quantum optimal control is performed iteratively. While the HQCA method is a good choice for large systems with uncertain

\[ \partial F/\partial \mathbf{u} \]  
(7)
Hamiltonians \cite{10,12}, it does not account for hardware transfer function. In case the bandwidth of the hardware transfer function is much narrower than the intended bandwidth of the inserted $\pi/2$ rotation pulse, the scheme can fail. Even if the $\pi/2$ pulse functions properly, HQCA measures $\partial F/\partial u$ instead of $\partial F/\partial \hat{u}$ (see Fig. 1), where $\hat{u}$ represents the distorted pulse. Compared to the radio-frequency regime typical of NMR, the microwave transfer function relevant to ESR experiments tends to be much less uniform over the frequency range of interest. Thus, we consider another method of closed-loop control that can take these effects into account: the FD method. It uses finite differences to approximate the derivative when an analytic expression of the gradient function is not available. If we write $u^i=(u_{z,A}, u_{y,A}, \ldots, u_{x,N}, u_{y,N})$ as the $q$th control amplitude parameters, $g^q$ can be expressed as

$$
 g^q = \sum_k g_k^i v^k, 1 \leq k \leq 2NM $$

$$
 g_k^q \approx \frac{F(u^i + \Delta u_k^q v^k) - F(u^i - \Delta u_k^q v^k)}{2\Delta u_k^q} \quad (8) $$

Here $\{v^k\}$ is a basis set that spans the parameter space and $\Delta u_k^q$ is a properly chosen difference value \cite{13}. To obtain the complete gradient vector $g^q$, similar to the HQCA method, a total of $4NMbp$ measurements are needed at each iteration. As the gradients are estimated directly from state fidelity measurements, distortions of the pulse due to the transfer function are accounted for in the process (see Appendix). This method is useful when the hardware transfer function is strongly frequency dependent or is not accurately known.

In Ref. \cite{13}, the authors simulated a closed-loop (in-situ) optimization scheme based on the FD method. Instead of using a complete basis set per iteration, they acquired gradient and performed optimization only with one random element of the basis set at a time. This results in fewer experiments per iteration, but convergence is slow if the random elements are not well chosen. In this work, we use two different basis sets: the first is a complete basis set in the time domain with a dimension of $2NM$ (we call it the linear basis), and the second is the Slepian basis \cite{22,24}. The Slepian basis can be constructed to have fewer elements than $2NM$ with narrower control bandwidth, and is thus suitable for applications when the bandwidth is experimentally limited (see Appendix).

**Experimental Results**— Experiments were carried out using a custom pulsed ESR spectrometer operating in X-band. An arbitrary waveform generation (AWG) enables pulse shaping, and a loop-gap resonator with $Q \sim 100$ allows excitation over a bandwidth $\sim 100$ MHz \cite{26}. The sample we use is a single crystal of unlabeled malonic acid (CH$_2$(COOH)$_2$), where paramagnetic defects are created by gamma-ray irradiation \cite{21}. Since the carbon atoms are not spin labeled, all hyperfine couplings involve surrounding hydrogen atoms (I=1/2), and up to 8 have been observed \cite{27}. The general spin Hamiltonian can be written as

$$
 H_0 = \mu_B B_0^T g \cdot S + \sum_{i=1}^{8} (S^T A^i \cdot I^i - \mu_n g_n B_0^T I^i), \quad (9) $$

where $\mu_B$ is the Bohr magneton, $\mu_n$ is the nuclear magneton, $B_0=B_0 \hat{z}$ is the externally applied magnetic field, $g$ is the $g$-tensor of the electron spin, $g_n$ is the $g$-factor of the nuclear spin, $S=(\hat{S}_x, \hat{S}_y, \hat{S}_z)$ is the electron spin operator, $A^i$ and $I^i$ are the hyperfine tensor and nuclear spin operator for
the \( i \)th nuclear spin, respectively (vectors are in bold). The hyperfine coupling to the \( \alpha \)-proton dominates, as it is about 10 times stronger than the second largest coupling. Therefore, we neglect all other protons and write a simplified, two-spin Hamiltonian:

\[
H_0 = \omega_1 I_z + A S_z I_z + B S_z I_x.
\]  

(10)

This Hamiltonian is written in the rotating frame of the electron and makes use of the secular approximation. Here \( A \) and \( B \) are the secular and pseudo-secular hyperfine couplings, respectively, and \( \omega_1 = \mu_{n g} B_0 \) is the nuclear Zeeman frequency. At X-band where the strength of the static magnetic field \( (B_0) \) is around 0.34 T for \( g \sim 2 \), \( |\omega_1| \sim 14.5 \text{ MHz} \). Diagonalizing \( H_0 \) gives

\[
H_0^d = \text{Diag} \left[ \frac{\omega_{12}}{2}, -\frac{\omega_{12}}{2}, \frac{\omega_{34}}{2}, -\frac{\omega_{34}}{2} \right]
\]

\[
|\omega_{12}| = \sqrt{(\omega_1 + A/2)^2 + B^2/4}
\]

\[
|\omega_{34}| = \sqrt{(\omega_1 - A/2)^2 + B^2/4},
\]  

(11)

where the superscript ‘\( d \)’ denotes the diagonal form. Figure 2(a) shows the energy level diagram for the hyperfine coupled electron-nuclear spin system. The hyperfine frequencies \( \omega_{12} \) and \( \omega_{34} \) are given in Eq. (11). The hyperfine coupling is known to be strongly anisotropic [26], so the values of \( A \) and \( B \) depend on how the sample crystal is oriented with respect to \( B_0 \). We chose an orientation in which \( A \gg B \). Under such condition, it is more difficult to fully characterize the Hamiltonian experimentally compared to the cases when \( A \sim B \). The uncertainty in the Hamiltonian provides a good testbed for comparing feedback control schemes.

As shown in Fig. 2(b), a spin echo sequence is utilized to read out the intensity of a particular ESR transition. When either the dc magnetic field, \( B_0 \), or the microwave frequency is varied to satisfy the resonance condition, strong peaks corresponding to the two allowed ESR transitions appear, as shown in Fig. 2(c). In addition, smaller signals from the forbidden transitions appear between the two strong allowed peaks.

From spectral fitting, the forbidden transition rates are estimated to be <5% of the allowed transition rates. The estimated range of \( A \) and \( B \) is 72>|\( A > 66 \text{ MHz} \) and 0<|\( B | < 26 \text{ MHz} \), where one constraint is that \( |\omega_{12}| = |\omega_{34}| = 72 \text{ MHz} \) (separation of the two allowed transitions). Since the forbidden transition rates are small, no electron spin echo envelope modulation (ESEEM) signals were observed. The lack of information from an ESEEM experiment is a key reason that the Hamiltonian parameters cannot be determined more accurately in this orientation.

Finally, we test and compare open- and closed-loop quantum optimal control of two state-to-state gates on the two-qubit system described above. We denote the thermal equilibrium state by \( \text{ZI} \), where the first (second) letter refers to the state of electron (nuclear) spin. Gate 1 is the transformation \( \text{ZI} \rightarrow \text{XI} \), and gate 2 is the transformation \( \text{ZI} \rightarrow \text{ZZ} \). Both target states only contain one Pauli element and thus \( P = 1 \). Characterizing the control quality requires measurement of the final state. This is done via two separate, selective readouts of the allowed ESR transitions, which we denote \( S_L \) and \( S_R \) (see Fig. 2(b)).

The minus sign in \( F_{ZZ} \) is due to the fact that the two allowed transition peaks have opposite sign in the ideal spectrum for the state \( \text{ZZ} \). We note that while \( F_{ZZ} \) can be considered as the true state fidelity, \( F_{XI} \) should be treated as a relative measure only. The reason is that \( F_{XI} \) can be larger than 1 when gate 1 performs better in exciting the transitions than the square \( \pi/2 \) pulse used in the reference measurement (see Fig. 2(b)).

---

**Figure 3.** (Color online) Process flow for optimizing closed-loop control. Steps are described in detail in the main text.

Step 1: Start with previous pulse

Step 2: For \( k = 1, k_{\text{max}} \), measure dummy \( F^k_{\text{XX}} \) and ideally \( R^k_{g}(\pm\pi/2) \)

Step 3: Compute gradients, \( g_x, F_{\text{XX}}, F_{\text{XX}} \)

Step 4: Sum over \( k \) to obtain final gradient \( g = \sum g_x \)

*Waveforms corresponding to different basis sets \( v \)*

FD (linear or Slepian)

HQCA

Linear

Slepian

Step 5: Update pulse by adding \( cg \) to \( x \)-phase. Similarly obtain gradient in \( y \)-phase.
by adding \( k \) to the number of pulse segments for the HQCA method, or from 1 to the size of the basis set \( v^k \) for the FD method.

**Step 3:** The \( x \)-phase gradient is given by \( g_k \propto F^{k,\pm x} - F^{k,-x} \) (see Eq. (7) for HQCA or Eq. (8) for the FD method).

**Step 4:** By summing over all \( k \), we obtain the full gradient for the current iteration in \( x \)-phase, \( g = \sum_k g_k v^k \). For the HQCA method, \( v^k \) is a unit vector with the only non-zero element being 1 at the \( k \)th index, e.g., \( \{0,1,0,...,0\} \) for \( k=3 \). For the FD method, it is the \( k \)th basis vector from a chosen set; in this paper we use linear and Slepian bases.

**Step 5:** We update the \( x \)-phase pulse from previous iteration by adding \( c g \). Here, \( c \) is a scaling factor chosen to avoid over- or undershooting. The \( y \)-phase pulse is updated in a similar manner, after finding the \( y \)-phase gradient.

Figure 4 summarizes the closed-loop optimization of the \( ZI \rightarrow ZZ \) pulse (gate 2). Similar to open-loop methods like GRAPE, the fidelity is seen to increase quickly in the first few iterations, but slows down and eventually saturates when the measured gradient becomes comparable to the shot noise. Although there is a convergence proof [18, 28] in case of noisy measurements, there are two problems in practice: (1) inaccuracy in measured gradients and (2) difficulty in verifying small improvements in \( F \). Moreover, a long-term drift in measurements can prevent \( F \) from reaching a convergence [18]. In practice, we found that there was no benefit in going beyond \( \sim 15 \) iterations for the gates and protocols tested here, i.e. when the improvement in \( F \) per iteration is smaller than the measurement noise.

Table I summarizes the final \( F_{X1} \) and \( F_{ZZ} \) obtained using three closed-loop optimal quantum control methods, (i) HQCA, (ii) FD with linear basis, and (iii) FD with Slepian basis. In addition, the results of open-loop control are presented. Here, the GRAPE pulses [25] were designed under three different conditions: (i) \( A=72 \text{ MHz}, B=0 \text{ MHz}, \tau=1 \), (ii) \( A=66 \text{ MHz}, B=26 \text{ MHz}, \tau=1 \), and (iii) \( A=66 \text{ MHz}, B=26 \text{ MHz}, \tau=\tau_{\text{meas}} \). \( \tau \) denotes the transfer function, and \( \tau_{\text{meas}} \) means that the pulses were designed under an ideal, flat transfer function. \( \tau\approx\tau_{\text{meas}} \) indicates that the pulse design accounted for the experimentally measured transfer function (see Appendix).

First of all, we remark again that direct comparison of \( F_{X1} \) and \( F_{ZZ} \) is not proper as \( F_{X1} \) is not strictly a fidelity. For open-loop quantum optimal control, we observe higher \( F \) when more accurate Hamiltonian parameters and realistic \( \tau \) are taken into consideration. However, closed-loop quantum optimal control methods still outperform the best open-loop results. Both closed-loop methods, HQCA and FD, produced similar control qualities under the experimental conditions tested here. However, simulations show that under different conditions, one method can perform better. This is described in the section below.

**Simulations**— Simulations were performed to further elucidate the roles of measurement noise and the spectrometer transfer function in limiting the final closed-loop control quality. In these simulations, the closed-loop optimization is performed in the same way as before, but with the experimental system response simulated by computer. The simulations were ended when the overall improvement after five successive iterations is smaller than 0.01. As shown in Table II, the simulation results indicate that HQCA is more robust to the measurement noise than FD methods. We find that this increased robustness for HQCA is due to its larger gradients compared to the FD methods.
Table I. Experimental control qualities for gate operations 1 and 2. For closed-loop control, results are given for HQCA, FD with linear basis, and FD with Slepian basis. For open-loop control, the Hamiltonian parameters were varied as well as whether or not the spectrometer transfer function is accounted for in pulse design. The numbers reported here are averages of 50 measurements, and given in the parentheses are standard deviations (e.g. $0.968(39)=0.968\pm 0.039$). \(^\dagger\) The open-loop control pulses were designed using the full-bandwidth basis set. * The open-loop control pulses were designed using the limited-bandwidth basis set, i.e. the Slepian basis, with a control bandwidth of 120 MHz. It should be noted that direct comparison of $F_{X1}$ and $F_{ZZ}$ is not proper as $F_{X1}$ is not strictly a fidelity (see the main text).

|            | Closed-loop control | Open-loop control |
|------------|---------------------|-------------------|
|            | HQCA  | FD (linear) | FD (Slepian) | $A=72\,\text{MHz}$, $B=0\,\text{MHz}$, $T=1$ | $A=66\,\text{MHz}$, $B=26\,\text{MHz}$, $T=1$ | $A=66\,\text{MHz}$, $B=26\,\text{MHz}$, $T=\text{meas}$ |
| $F_{X1}$   | 0.968(39) | 0.993(46) | 1.010(40) | 0.946(40) \(^\dagger\) 0.951(41) \(^*\) 0.955(37) \(^\dagger\) 0.990(51) \(^*\) |
| $F_{ZZ}$   | 0.914(36) | 0.918(43) | 0.932(37) | 0.807(37) | 0.807(37) \(^*\) 0.799(37) \(^*\) 0.891(40) \(^\dagger\) 0.883(45) \(^*\) 0.889(39) \(^\dagger\) 0.902(43) \(^*\) |

Table II. Simulation results showing the effects of noise level and transfer function bandwidth on the control fidelities $F_{ZZ}$ for gate 2. Noise: Zero-mean Gaussian random noise with a standard deviation ($\sigma$) of 0.03, 0.07, 0.14, and 0.20 was added to the ideal gradient measurements, where the reference measurements ($\overline{S}_R$ and $\overline{S}_L$) are normalized to 1. The averages and standard deviations of ten trials in each condition are given. Transfer function: In each case, the entire control pulse sequence was distorted according to a realistic transfer function with full width at half maximum (FWHM) of 130 and 70 MHz. In the simulation with different noise levels, the transfer function with FWHM of 130 MHz FWHM was considered. Simulated pulse finding was stopped when the overall improvement over five successive iterations was smaller than 1%. $T_2$ was not included in this simulation.

| Noise | HQCA | FD (linear) | FD (Slepian) |
|-------|------|-------------|--------------|
| ~0.03 | 0.958(02) | 0.967(01) | 0.973(02) |
| ~0.07 | 0.958(02) | 0.968(04) | 0.973(03) |
| ~0.14 | 0.957(03) | 0.947(26) | 0.960(30) |
| ~0.20 | 0.956(09) | 0.926(48) | 0.905(29) |

| FWHM  | HQCA | FD (linear) | FD (Slepian) |
|-------|------|-------------|--------------|
| ~130MHz | 0.958  | 0.970  | 0.975   |
| ~70MHz  | 0.936  | 0.974  | 0.964   |

Conclusions—Two closed-loop quantum optimal control methods are experimentally demonstrated in a solid-state two-qubit system. Together with simulations, we find that the closed-loop quantum optimal control methods outperform the open-loop quantum optimal control method when the information of the Hamiltonian and hardware transfer function lacks accuracy. HQCA works better than FD methods when shot noise in measurements is large enough to be the dominant error limiting the gradient search. When $T$ is narrow and/or the control bandwidth is limited (often by hardware), FD methods can perform better than HQCA. With the bandwidth-limited Slepian basis set, the gradient finding procedure of the FD method can be made less time-consuming and the pulses generated may be friendlier to implement in experiment. Thus, starting with a viable open-loop quantum optimal control (e.g. GRAPE) pulse and running subsequent iterations of an appropriate closed-loop control protocol may be a good strategy to reach high control quality under realistic experimental conditions. In the future, it may be of interest to combine the optimal random orientation method [28, 29] with the closed-loop quantum optimal control methods for better efficiency of convergence rate.

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Appendix A: Difference between the gradient derived with the HQCA and FD methods

As mentioned in the main text, HQCA does not consider the existence of hardware transfer function. In case of an extremely narrow transfer function, the required $\pm \pi/2$ pulses can be seriously distorted and the method can totally fail. When the distortion is mild, the $\pm \pi/2$ pulses can still function well, which is the case for our experiment. Here we describe the difference between the gradient derived with the HQCA and FD methods.

The target function, which is the state fidelity $F$, is a function of the control amplitudes $u$, i.e. $F(u)$. In presence of hardware transfer function $\mathcal{T}$, it can be further written as $F(\tilde{u})=F(\mathcal{T}(u))$ where $\tilde{u}$ is the distorted control amplitudes. In HQCA, the derived gradient is

$$\tilde{g} = \frac{\partial F}{\partial u}.$$  \hspace{1cm} (A1)

In the FD method, the derived gradient is $$g = \frac{\partial F}{\partial u}.$$  \hspace{1cm} (A2)

The relationship between $\tilde{g}$ and $g$ is

$$g = \tilde{g} \cdot \frac{\partial \tilde{u}}{\partial u},$$  \hspace{1cm} (A3)

where $\frac{\partial \tilde{u}}{\partial u}$ is given by $\mathcal{T}$. The gradient $g$ is the “correct” gradient for updating the control amplitudes $u$. As shown with the simulation in the main text, narrower bandwidth of $\mathcal{T}$ yields lower final fidelity with HQCA while fidelities obtained using the FD method are practically independent of $\mathcal{T}$. This indicates that the gradient derived with the FD method automatically accounts for $\mathcal{T}$. If $\mathcal{T}$ is known accurately, obtaining $g$ from $\tilde{g}$ is possible using Eq. (A3).

Appendix B: Non-ideal $\pm \pi/2$ rotation pulse in HQCA

The pulse we insert in HQCA should ideally be a non-selective $\pm \pi/2$ rotation, but this is difficult to realize with simple square or Gaussian pulses due to the limited control bandwidth and pulse power. So we try to realize the rotation by simply adding two selective $\pm \pi/2$ square pulses at the two different frequencies, given by the distance between the two allowed ESR peaks from the center (i.e. $\pm 36$ MHz; see Fig. 2(c) of the main text). Even though the composed pulse does not provide accurate $\pm \pi/2$ rotation, the simulation and experiment confirm that it is sufficient in deriving gradient toward higher control quality. Here, we give a short proof for the effectiveness of a non-ideal $\pm \pi/2$ rotation in the HQCA method. From Ref. [10], we have

$$\frac{\partial F}{\partial u_{\alpha,l}(m)} = \frac{\text{Tr}(-i\Delta t U_{m+1}^{\dagger}[\sigma^l_{\alpha,l} U_{1}^{\dagger}U_{1}^{\dagger}]U_{m+1}^{\dagger}\rho)}{2^n},$$  \hspace{1cm} (B1)

and for any operator $\rho$,

$$[\sigma^l_{\alpha}, \rho] = i[R^l_{\alpha} (\frac{\pi}{2}) \rho R^l_{\alpha} (\frac{\pi}{2}) ] - R^l_{\alpha} (-\frac{\pi}{2}) \rho R^l_{\alpha} (-\frac{\pi}{2})].$$  \hspace{1cm} (B2)

By combining Eqs. (B1) and (B2), we obtain Eq. (7) in the main text. When the rotation angle $\theta \neq \pi/2$, it is easy to see that

$$[\sigma^l_{\alpha}, \rho] = \frac{i[R^l_{\alpha} (\theta) \rho R^l_{\alpha} (\theta) ] - R^l_{\alpha} (-\theta) \rho R^l_{\alpha} (-\theta)]}{\sin(\theta)},$$  \hspace{1cm} (B3)

meaning that the orientation of the derived gradient is unaffected (the amplitude is scaled by a factor of $\sin(\theta)$).

Appendix C: Bandwidth-limited Slepian basis

The Slepian basis set we use in this work is composed of Slepian sequences which are also called discrete prolate spheroidal sequences [24]. Slepian sequences with a sequence length $N$ and half bandwidth $W \in (0, 0.5]$ are defined to be the real solutions to the eigenvalue problem

$$\sum_{m=0}^{N-1} \frac{\sin(2\pi W(l-m))}{\pi(l-m)} v_k(m;N,W) = \lambda_k(N,W) v_k(l;N,W).$$  \hspace{1cm} (C1)

Here, $k, l \in \{0, 1, ..., N-1\}$, and $v_k(l; N, W)$ is the $l$th element of the $k$th order Slepian sequence. The Slepian sequences are spectrally concentrated in the frequency range $[-W/\Delta t, W/\Delta t]$, especially the first $2NW$ ones. Ref. [23] used the first $2NW$ Slepian sequences to approximate the space of bandwidth limited sequences of length $N$.

Appendix D: Linear basis

The shaped pulse used in this work has 100 segments with 2 ns step size. In addition to the bandwidth-limited Slepian basis described above, a linear basis set of dimension 100 based on the Hadamard basis is used. The complete space of this linear basis set is composed of three subspaces with 64-dimensional, 32-dimensional and 4-dimensional Hadamard basis spaces. The linear basis set is chosen this way because having as many non-zero elements as possible in the basis set is preferred to generate observable difference in the FD method.
Appendix E: Transfer function

Our hardware transfer function $T$ was measured by observing the Rabi oscillation of the electron spin signal at different offset frequency of the control pulse (see Ref. [9] for more detail). By fitting the oscillation trace, the response of the spin to the pulse with different frequency can be obtained as shown in Fig. S1(a). This was used in the open-loop control pulse design and simulation. The transfer function in Fig. S1(b) is an artificial alteration from Fig. S1(a) to make the bandwidth narrower for simulation purpose only.

Appendix F: Effect of additional H spins in the environment

Throughout the paper, we use the simplified, two-spin Hamiltonian with one electron and one $\alpha$-H spin. However, there are more H spins in the environment that are weakly coupled to the electron spin (Ref. [27] identifies seven more H spins with an order of magnitude smaller coupling strengths compared to the $\alpha$-H spin). Such weak couplings are not resolvable in the ESR spectrum because the linewidth is too broad ($\sim$10 MHz; see Fig. 2(c) of the main text). And we treated the effect of the weak environment H spins as a static inhomogeneous local field. However, these environment H spins will likely change their state during our control pulse. In order to gain some insight about the influence of additional environment H spins on our closed-loop optimal control, further simulation of ZI-ZZ gate using the FD method was performed with one electron, one $\alpha$-H, and one weakly coupled H spin. For the weakly coupled H spin, different coupling strength constants of $A$ and $B$ were considered. It was found that while the strength of $A$ has little effect, larger $B$ value tends to slow down the process. For example, the simulation with $A=4$ MHz and $B=0$ MHz generated the same fidelity value compared to the simulation without the weakly coupled H, under same termination condition (e.g. when the average fidelity improvement is less than 0.002 per iteration). However, when $A=0$ MHz and $B=4$ MHz, the final fidelity was 0.004 lower than the simulation without the weakly coupled H. Therefore, the environment H spins can be another error source, especially if they have non-zero $B$. Due to large amount of computing resources required and uncertainty in the crystal orientation, we did not simulate all the seven H spins identified by Ref. [27]. The effect of weakly coupled environment H spins on closed-loop optimal control is of future interest.