Strong driving of a single spin using arbitrarily polarized fields

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The strong driving regime occurs when a quantum two-level system is driven with an external field whose amplitude is greater or equal to the energy splitting between the system’s states, and is typically identified with the breaking of the rotating wave approximation (RWA). We report an experimental study, in which the spin of a single nitrogen-vacancy (NV) center in diamond is strongly driven with microwave (MW) fields of arbitrary polarization. We measure the NV center spin dynamics beyond the RWA, and characterize the limitations of this technique for generating high-fidelity quantum gates. Using circularly polarized MW fields, the NV spin can be harmonically driven in its rotating frame regardless of the field amplitude, thus allowing rotations around arbitrary axes. Our approach can effectively remove the RWA limit in quantum-sensing schemes, and assist in increasing the number of operations in QIP protocols.

I. INTRODUCTION

The nitrogen-vacancy (NV) center is one of the leading platforms for QIP applications⁴⁶,⁸,¹⁴ and room-temperature quantum metrology⁵,⁶. Additionally, it serves as a probe for the classical and quantum dynamics of a mesoscopic bath of spins⁰¹. These applications provide great motivation for controlling, and specifically shortening the manipulation duration of the spin. In QIP, shortening of the gate duration allows an increase in the number of quantum-gates applied during the coherence time, T₂, and thus scales up the computational performance.⁴ Ultimate, the elementary gate duration defines the processing clock-speed, for systems on parity for the number-of-operations figure of merit, it distinguishes between “slow” systems, such as cold trapped atoms or nuclear spins, and “fast” systems such as semiconductor quantum dots and superconducting flux qubits. In quantum metrology, designed rotations of qubits are used to map the signal (the phase between eigenstates) to a measurable population difference. Specifically, in dynamical decoupling (DD) based quantum sensing schemes, the qubit may be driven continuously or pulsed at intervals, allowing suppression of noise sources with a slower spectrum than the driving speed/interpulse spacing⁸¹.¹⁶ Thus, the maximum driving speed or pulse duration places an upper bound on the ability to shift the sensing frequencies higher and away from the dominant low frequency noise, and limits the bandwidth of these schemes.

For these goals, and crucially in room temperature applications, the NV spin is usually manipulated with an oscillatory microwave (MW) field Bₓ (t) = B₁ cos (ω₁ t), (where B₁, ω are the field amplitude and frequency respectively), resonant with the energy splitting of the spin hω₁, i.e. ω = ω₁. Then, in a frame rotating with the MW field, the spin is driven by a constant magnetic field B₁ = B₁/2 (co-rotating field), and an additional rotating field B₂ = (B₁/2) e⁻iω₁t (counter-rotating field). As long as γB₁ is small compared to ω (where γ is the magnetic moment of the spin), the counter-rotating field can be neglected, an approximation known as the rotating wave approximation (RWA). In this regime, the gate time depends linearly on the inverse of applied magnetic field amplitude B₁.

However, when the driving amplitude is increased so that γB₁ becomes comparable to the spin’s Larmor frequency ω₁, the dynamics manifest complexities due to an interplay of the two fields: the gate fidelity degrades, the rotation (gate) time no longer scales linearly with 1/B₁, and the dynamics show pronounced sensitivity to the phase of B₁(t) with respect to the pulse edge.⁸² In this regime, known as the strong driving regime, various solutions to regaining control of the system dynamics have been proposed, including anharmonic pulses¹⁵,¹⁶, Landau-Zener assisted transitions, and transitions through an ancillary level in a Λ-type configuration.

Here we tackle the strong driving problem using an approach discussed in an early work of Bloch and Siegert.⁴ A spin subjected to two orthogonal, resonant MW fields Bₓ = B₁ cos (ω₁ t) and Bᵧ = B₁ cos (ω₁ t + φ) will rotate harmonically if φ = ±π/2. Under this condition, the two orthogonal fields can equivalently be described as circularly polarized MW radiation. When the radiation polarization coincides with the spin transition (i.e. when the angular momentum of the radiation field matches the change in spin number), manipulation with a field solely co-rotating with the spin occurs, leading to full contrast rotations. The other case, namely driving a transition with the counter-rotating field (of opposite handedness/polarization), can be viewed as a driving field with a 2ω₁ detuning. Only in the strong driving regime, may rotation of the spin occur, albeit with degraded contrast. Recently, this approach was demonstrated with an ensemble of ¹H nuclear spins (I = 1/2) in an ultra-low field NMR setup. Here, we investigate this approach using experiments on a single NV center, an electronic spin in diamond with total spin S = 1. For an S = 1 system, it is possible to address more than one transition spectrally, enabling polarization selective transitions.¹⁷ We drive the NV center with arbitrarily polarized MW radiation, address one of the two-level systems, and study its dynamics in the strong driving regime, namely, when
the Rabi frequency is larger than the Larmor frequency.

The paper is organized as follows. In Section II we describe the experimental setup, and present a theoretical description of the general Hamiltonian of the NV $S = 1$ ground-state under two MW fields. Section III discusses the dynamics in low magnetic field, characterized by selective excitation within a dense spectrum of resonances. In Section IV we experimentally demonstrate the strong driving regime for various polarizations, and compare between the dynamics under linear and circular polarizations. In Section V we discuss the results and elaborate on the effect of an axial MW component on the NV dynamics, i.e a MW field applied parallel to the NV dipole axis.

II. EXPERIMENTAL SETUP FOR POLARIZED MW RADIATION

The experiments were conducted at room temperature, with single NV centers in a type IIa diamond with (100) surface. To excite NV centers with arbitrarily polarized MW pulses of short duration, we designed a low-Q MW antenna. The antenna comprises of two thin copper wires in a cross-configuration, stretched over the diamond surface (Fig. 1a). The wires were connected to two independent MW sources, switches, and amplifiers, and were phase-locked to each other. Alternatively, one can apply the fields through an arbitrary waveform generator to gain full control over the MW parameters. With this setup we were able to manipulate individual NV centers, located at distances of ~10-50μm from the wire crossing, with Rabi frequencies up to 100 MHz. We have found that the position of the NV center with respect to the wires affects the driving performance (See Sec.V). The ideal scenario is illustrated in Fig. 1a, where the two fields and the NV axis form an orthogonal system.

The Hamiltonian of the NV center spin, $\hat{S}$, in the presence of two orthogonal driving fields $B_x, B_y$ of equal magnitude, and a constant external magnetic field $B_{ext}$, can be written as

$$H = D S_z^2 - \gamma B_{ext} S_z + \Omega e^{i(\omega t + \phi)} \hat{\varepsilon} \cdot \hat{S} + \text{h.c.},$$

where $D = (2\pi)2.87$ GHz is the zero-field splitting, $\gamma = (2\pi)2.8$ MHz/G is the NV magnetic moment, $\hat{\varepsilon} = (1, e^{i\theta}, 0)$ defines the MW polarization and h.c. stands for hermitian conjugate. Here, $\omega, \Omega$, and $\phi$ are the MW frequency, the NV Rabi frequency ($\Omega = \gamma B_x = \gamma B_y$), and relative MW phase, respectively. The phase $\phi$ is a global phase shared by both fields. In the rotating frame, Eq. (1) is rewritten as (See Appendix A)

$$H' = \begin{bmatrix}
\Delta_+ & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & \Delta_-
\end{bmatrix} + \frac{\Omega}{\sqrt{2}} \begin{bmatrix}
0 & \varepsilon_- & 0 \\
\varepsilon_+ & 0 & \varepsilon_+ \\
0 & \varepsilon_- & 0
\end{bmatrix},$$

where $\varepsilon_{\pm} = e^{\mp i\phi} \left(1 - i e^{\mp i\phi}/2\right)$. Here $\omega = \gamma B_{ext}$ are the transition frequencies, and $\Delta_{\pm} = \omega_{L} \mp \omega$ their detuning from the microwave frequency (Fig. 1b). The second and the third terms of Eq. (2) represent the counter-rotating and rotating terms, respectively. Note that Eqs. (2) hold for arbitrarily (elliptically) polarized fields, however we assume that the z-component of the MW field is zero. We refine this treatment in Section V, when discussing the influence of a MW field with non-zero axial component.

Next we discuss the dynamics of Eq. (2) in the ‘low field’ case, where the transitions are nearly degenerate with respect to the Rabi frequency, but the RWA is applicable ($\Delta_{\pm} \ll \Omega \ll \omega$). Then, we discuss the dynamics in the ‘high-field’ case where the transitions are well...
separated and the Rabi frequency exceeds the transition frequency \((\Delta \ll \omega \ll \Omega)\), allowing investigation of a two-level system driven beyond the RWA limit.

III. SELECTIVE EXCITATION WITH POLARIZED FIELDS

To characterize the performance of the MW structure we performed broadband Ramsey magnetometry at low magnetic field, where the \(|1\rangle\) and \(|+1\rangle\) states are nearly degenerate. For low amplitude driving \((\Omega \ll \omega)\) one may use the RWA, i.e. assume that the \(\epsilon_{\pm}|e^{\pm 2i\omega t}\) components oscillate many times during the duration of the spin, and thus are averaged to zero. Eq. \((2)\) then becomes

\[
H_{\text{RWA}}^{\phi} = \begin{pmatrix}
\Delta_- & (\Omega/\sqrt{2}) \epsilon_-^* & 0 \\
(\Omega/\sqrt{2}) \epsilon_+ & 0 & (\Omega/\sqrt{2}) \epsilon_+^* \\
0 & (\Omega/\sqrt{2}) \epsilon_-^* & \Delta_+
\end{pmatrix}.
\]

(3)

Here we see that \((\Omega/\sqrt{2}) \epsilon_-\) drives the \(|0\rangle \leftrightarrow |+1\rangle\) transition, and \((\Omega/\sqrt{2}) \epsilon_-\) drives the \(|0\rangle \leftrightarrow |-1\rangle\) transition.

The NV is first optically pumped to the \(|0\rangle\) state. Then, a MW \(\pi/2\)-pulse with arbitrary polarization (arbitrary \(\phi\)) manipulates the NV spin to the state \(|\psi\rangle = (1/\sqrt{2}) \left[ |0\rangle + e^{i\phi} |1\rangle + e^{-i\phi} |-1\rangle \right]\). This state can be obtained with the evolution operator \(U = \exp(iH_{\text{RWA}}^{\phi} t)\), for rotation time \(t\) satisfying \((t \cdot \Omega/\sqrt{2}) = \pi/2\), in the limit \(|\phi| \gg \Delta_\pm\). Then, after a free-evolution time, \(\tau\), the state becomes \(|\psi\rangle = (1/\sqrt{2}) \left[ |0\rangle + e^{i\phi} e^{i\Delta_+ \tau} |1\rangle + e^{-i\phi} e^{-i\Delta_- \tau} |-1\rangle \right]\), and an additional \(\pi/2\)-pulse with the same polarization gives the final probability to be in the \(|0\rangle\) state as

\[
P_0(\tau) = \frac{1}{2} \left[ 1 - |\epsilon_-|^2 \sin(\Delta_\pm \tau) - |\epsilon_+|^2 \sin(\Delta_\pm \tau) - |\epsilon_-|^2 |\epsilon_+|^2 (\cos((\Delta_+ - \Delta_-) \tau) - 1) \right].
\]

(4)

The second and third terms oscillate at the microwave frequency detuning from the \(|0\rangle \leftrightarrow |-1\rangle\) and \(|0\rangle \leftrightarrow |+1\rangle\) transitions, respectively. The last term oscillates at the frequency separation between the \(|\pm\rangle\) states, and is detuning independent. Using Fourier analysis of \(P_0(\tau)\), one can infer the polarization parameters \(||\epsilon_{\pm}|^2\rangle\) directly, by observing the intensity of each frequency component.

In the experiments, a static axial magnetic field of 4.6 Gauss was used to split the \(|\pm\rangle\) states by 26 MHz, and \(\pi/2\)-pulses which efficiently excited both transitions were applied \((\Omega = (2\pi)114\text{MHz})\), note that \(\Omega\) remained still much smaller than \(\omega_L \approx (2\pi)3 \text{ GHz}\). By varying the relative phase between the wires, various polarizations could be engineered; left-handed circular polarization (driving the \(|0\rangle \rightarrow |-1\rangle\) transition, Fig. \((11)\)), right-handed circular polarization (driving the \(|0\rangle \rightarrow |+1\rangle\) transition, Fig. \((9)\)), and linear polarization (Fig. \((12)\)). In all spectra there is an additional \((2\pi)2.16 \text{MHz}\) splitting due to hyperfine interaction with the hydrogen nuclear spin from the relative amplitudes in the spectral footprint, we deduce that \(|\epsilon_-|^2 = 0.98, 0.03, 0.47\) (with an error of \(\pm 0.05\)) for Figs. \((11)\), \((12)\), and \((14)\), respectively.

IV. STRONG DRIVING WITH ARBITRARY POLARIZATIONS

After characterizing the MW polarization, we experimentally investigated the strong driving regime for different applied polarizations. A two-level system (TLS) was prepared using a high axial magnetic field \(\sim 0.1 \text{ mT}\). At this field one finds \(\omega_L = (2\pi)30 \text{MHz}\), and \(\Omega = (2\pi)5710 \text{MHz}\). For a field resonant with \(\omega_L\), the far-detuned higher energy transition can be neglected, and the reduced Hamiltonian of the two-level system \(|0\rangle, |1\rangle\) derived from Eq. \((2)\) is

\[
H = \frac{\Omega}{\sqrt{2}} \begin{pmatrix}
0 & e^{2i\omega_L t} \\
e^{-2i\omega_L t} & 0
\end{pmatrix}
\]

(5)

where \((\Omega/\sqrt{2}) \epsilon_-\) is the co-rotating component of the MW field, and \((\Omega/\sqrt{2}) \epsilon_+ e^{-2i\omega_L t}\) is the counter-rotating component. Specifically, for \(\phi = \pi/2\), one obtains the Hamiltonian \(H = (\Omega/\sqrt{2}) \sigma_x\), where \(\sigma_x\) is the Pauli matrix, and the prefactor \(\sqrt{2}\) is a remnant of the \(S = 1\) nature of the NV system (The spin interacts stronger than a true TLS). This Hamiltonian is exact, and independent of the driving field magnitude, even for \(\Omega \gg \omega_L\), i.e. beyond the RWA limit. The dynamics derived from this Hamiltonian are harmonic oscillations with Rabi frequency \(\Omega\). We note that our experiments were conducted with Rabi frequencies on the order of tens of MHz, but in principle could be performed at the GHz regime with the proper hardware. The approximation leading from Eq. \((2)\) to Eq. \((5)\) breaks only at \(\Omega \sim 2D \simeq (2\pi)6 \text{GHz}\). For these values parasitic excitations to the \(|+1\rangle\) state will interfere with the dynamics.

A. Optimization of the relative phase \(\phi\)

As described above, experimental control of the MW polarization is obtained by tuning the relative phase, \(\phi\), between the wires. To further illustrate this control, we performed Rabi oscillations for various relative phases (Fig. \((2)\)). We started with a parameric scan in the weak driving regime. The NV spin was driven with both wires, each with amplitude \(\Omega = 0.15\omega_L\) \(( (2\pi)5.9 \text{MHz})\), and the relative phase between the sources was scanned (Fig. \((2)\)). At the optimal phase relation, \(\phi = \pi/2\), the NV spin is driven most efficiently, resulting in Rabi oscillations at double the frequency \(0.3\omega_L\) (Fig. \((2)\), red dashed line), corresponding to driving with the co-rotating field, and the counter-rotating term is suppressed completely \((\epsilon_- = 1, \epsilon_+ = 0)\). In contrast, at \(\phi = 3\pi/2\) the spin remained untouched (Fig. \((2)\), green dotted line), as
the MW has the opposite polarization to drive the spin transition. Here, the co-rotating field does not exist ($\varepsilon_- = 0, \varepsilon_+ = 1$), and the counter-rotating field can be neglected via the RWA ($\Omega \ll \omega_L^{-1}$). In contrast, in the strong driving regime the spin is also driven by the counter-rotating field (Fig. 2b). Here, we set $\Omega = 0.7\omega_L$ for each wire, and only at $\phi = \pi/2$ (representing left-handed circular polarization, $\sigma^-$) pure harmonic oscillations were observed, demonstrating Rabi flops with $\Omega = 1.4\omega_L^{-1}$. Hereafter, we denote the ratio of the Rabi frequency to the Larmor frequency as $\lambda = \Omega/\omega_L$. For other phases, more complex dynamics were observed, accompanied with high frequency components and lower contrast, specifically, at $\phi > \pi$, the dynamics are governed by the counter-propagating field and one notices an increase in the oscillation frequency with low contrast.

For both cases, a numerical model based on Eq. (5) reproduces the results very well. For all non-circular polarizations, the phase of the MW with respect to the pulse rising edge has an important role in the dynamics. For example, with linearly polarized MW radiation, the effective driving field is $\tilde{\Omega} = \sqrt{2}\Omega e^{-i(\omega_L^{-1} t - \pi/4)} \cos(\omega_L^{-1} t - \phi_g)$, representing a field with time-dependent magnitude and orientation. Assuming a square pulse shape (the rise and fall times of the experimental pulses were $\sim 1$ns), one finds that for $\phi_g = 0$, the field is $\tilde{\Omega} = \sqrt{2}\Omega \cos(\omega_L^{-1} t)$, and both fields (co- and counter-) start with maximal amplitude in the same direction (in the rotating frame), effectively rotating the spin instantaneously. In contrast, for $\phi_g = \pi/2$, the effective field is $\tilde{\Omega} = \sqrt{2}\Omega \sin(\omega_L^{-1} t)$. Here, the field has zero amplitude at $t = 0$, the spin starts to rotate much slower, drawing a different trajectory on the Bloch sphere. Conventionally, and in our experiments too, the trigger of the MW switch is not synchronized with the MW source phase, leading to a randomized initial phase $\phi_g$ over all acquisitions (each sequence was repeated $\sim 10^5$ times for sufficient photon statistics). Therefore, the simulated signal plotted in Fig. 2 is the averaged signal of 300 repetitions of the dynamics under Eq. (5) with uniformly distributed, global phases. In the weak driving regime the initial microwave phase is unimportant and the repeated acquisitions are essentially identical. For more details on the global phase dependence, see Appendix B.

B. Strong driving with linear and circular fields

After optimizing the relative phase for circular polarization ($\phi = \pi/2$) and for linear polarization ($\phi = 0$), we compare the performance of the two polarizations for manipulating the spin in the strong driving regime. Specifically, we compare the ability to steer the spin from the north pole of the Bloch sphere, $|0\rangle$ to the south pole, $|-1\rangle$, i.e. to perform a $\pi$ pulse.

Fig. 3a, b, and c, show the spin dynamics for $\lambda \sim 0.5, 1.0$ and 1.5, respectively. A qualitative difference is observed in the spin dynamics as the driving field exceeds the Larmor frequency; the oscillations become anhar-
monic for linear fields whilst remaining harmonic for circular fields. We extract two quantities from the measured signals: the time of the first minimum of the signal, $t_m$, and the $|0\rangle$-state population at this time. The former corresponds to a $\pi$-pulse duration for ideal harmonic driving, and the latter corresponds to the $\pi$-pulse fidelity, i.e. how well the spin is transferred from the $|0\rangle$ state to the $|−1\rangle$ state.

Fig. 3 shows the fidelity of $\pi$-pulse as a function of the effective Rabi strength (defined as half of the inverse of the $\pi$-pulse duration, i.e. $\Omega_{eff} = 1/2t_m$). For linear polarization the $\pi$-pulse fidelity decreases substantially when $\lambda \geq 1$ (Fig. 3d, rectangles), as predicted by a model based on Schrödinger equation with Eq. (5) (Fig. 3d, solid line). In contrast, for circular polarization the fidelity is 93% at $\lambda = 1$. Importantly, harmonic behavior of the driven spin is still observed for a Rabi frequency of twice the Larmor frequency. In principle the fidelity shouldn’t decrease even in the strong driving regime, however, for high $\lambda$-values the experimental values show monotonic reduction in the $\pi$-pulse fidelity. This behavior can be partly reproduced by simulations, if an additional field which is applied parallel to the NV-axis is included. This is illustrated by the five dashed lines in Fig. 3d, for which we added to Eq. (5) an additional term $H_\parallel = S_z\Omega_z \cos(\omega_L t)$, where $S_z$ is the spin operator in the $z$-axis, and $\Omega_z$ is the MW projection on the $z$-axis. The improved agreement between experiment and simulation for values of $\Omega_z/\Omega = 20\%-30\%$, implies that this could be a dominant mechanism for the degraded performance of circularly polarized radiation as the field amplitude is increased.

An additional figure of merit for the manipulation performance is the how the driving speed changes with the applied microwave amplitude (Fig. 3e). Here, for a linearly polarized MW field at $\lambda > 1.2$ multiple minima appear in the fluorescence signal (see Appendix B). As a consequence, the time of the first minimum changes abruptly at these values, shifting from the predicted behavior of $\Omega = \Omega_{eff} = 1/2t_m$ to higher values (Fig. 3d, rectangles), in agreement with our numerical model (Fig. 3d, solid line) the ideal behavior is depicted as a dotted black line. In contrast, for circular polarization the $\pi$-pulse duration follows the ideal behavior (Fig. 3b, circles), with a small deviation towards higher values. Again, this is partly explained by including an axial field (red dashed line, calculated with $\Omega_z/\Omega = 20\%$).

V. DISCUSSION

The comparison of the experiments and simulations in Fig. 3 indicates that an axial MW field could have an important role in our driving scheme. To verify this effect, we measured NV centers at various positions relative to the cross-wires, and selected an NV center with high axial component of the MW field. A scan of Rabi flops as a function of the relative phase between the wires is shown in Fig. 4. Here, without an axial component of the MW field, one would expect to obtain the results in Fig. 4a, where at $0^\circ$ (representing $\sigma^-$ polarization) the spin is driven with harmonic oscillations by the co-rotating field. In contrast, the experiment shows a qualitatively different behavior, where for phases in the range $0 - 90^\circ$, the oscillation frequency remains relatively constant, and the shape is clearly anharmonic (the blue solid line in Fig. 4 is a guide for the eye). Moreover, at the cancellation point ($180^\circ$), the spin is still rotated with Rabi frequency about fifth of the applied $\Omega$ (Fig. 4b). Remarkably, a numerical simulation based on Eq. (5) with axial component $S_z\Omega_z$, reproduces these features, elucidating the impor-
tance of axial driving for this NV center (Fig. 4b). At the cancellation point, for instance, the spin is likely to be driven via multiple Landau-Zener transitions, rather than with conventional Rabi flops.

Thus, the performance of our current design of a polarized MW antenna (cross-wires configuration) in the strong driving regime, is sensitive to the projection of the MW field on the NV center axis. Although axial driving is important for realizing Landau-Zener like transitions, it is also accompanied with a reduction of the oscillation contrast.

VI. CONCLUSION AND OUTLOOK

In conclusion, we studied the dynamics of a single spin under resonant, polarized MW radiation. The relative phase of two MW sources was utilized as a knob to adjust the MW polarization. We demonstrated high fidelity selective excitation within a dense spectrum of resonances, allowing individual excitation of adjacent transitions (Δ ~ 26 MHz) with fast pulses of Ω ≈ 114 MHz. This is of importance near level crossings, where conventionally one would have to decrease the driving power to avoid leakage of population to neighboring states, or use optimal control solutions. Here, the selection rules obtained with circularly polarized light allow selective excitation of degenerate transitions and can be used to determine both the sign and magnitude of the external magnetic field. We showed that under circular MW fields, the spin experiences pure harmonic oscillations regardless of the applied field strength, and specifically even above the RWA limit (in our case more than twice the Larmor frequency). Importantly, although being in the strong driving regime, the spin is still rotated in its rotating frame, allowing for universal control around the Bloch sphere by controlling the global phase of the MW fields ϕg. This enables the use of complex dynamical decoupling scheme with sub-Larmor period intrapulse duration. Moreover, in continuous wave sensing schemes such as dressed-state magnetometry and Hartmann-Hahn double resonance, the spin must be maintained in its dressed state. Here, our scheme overcomes the upper limit to detection frequencies set by the Larmor frequency.

Our current design suffers from the influence of an axial component of the MW field. More versatile structures, for example using two wires for generating each magnetic field component of the MW field, could mitigate this problem by allowing cancellation of the axial field without suppressing the transverse component. Spin manipulation with an axial field is strongly connected to Landau-Zener transitions and coherent destruction of tunneling, and is therefore interesting in and of itself. Moreover, our ability to control the magnetic field in all three direction can assist in constructing the Berry Hamiltonian, for acquiring controlled geometric phases with a single spin in diamond without rotating the sample.

Acknowledgments

The authors thank Jochen Scheuer, Xi Kong, and Christoph Miêjeller for assistance with experiments. The authors are grateful to Philip Hemmer, David Gershoni, Ran Fischer and Chen Avinadav for fruitful discussions and suggestions. The research was supported by DARPA, EU (ERC Synergy grant BioQ, DIAMANT), DFG (SFB TR 21, FOR 1493, FOR 1482), RSF, the Alexander von Humboldt and Volkswagen foundations.

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Figure 4: Strong driving of an NV center with large axial MW field (color online). Simulation of Rabi oscillations ($\omega_L = (2\pi) \times 37.8$ MHz, $\Omega_x = \Omega_y = 1.408 \pi \omega_L$) with (a) $\Omega_z = 0$ and (b) $\Omega_z = 1.67 \Omega_L$. (c) Experimental Rabi oscillations for a single NV center with $\omega_L$ and $\Omega_{x,y}$ equal to the values in the simulations, and with strong axial component.

We we derive the MW terms in the Hamiltonian of Eq.(2). The interaction term of the NV spin $S$, with MW field of frequency $\omega$, Rabi frequency $\Omega$, and relative phase $\phi$, is

$$H_{MW} = \Omega (\cos (\omega t) S_x + \cos (\omega t + \phi) S_y)$$

where the transverse spin operators are

$$S_x = \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \quad S_y = \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & -i & 0 \\ i & 0 & -i \\ 0 & i & 0 \end{pmatrix}.$$

We move into the MW rotating frame using the transformation $H' = i \frac{dH}{dt} U^{-1} - U^\dagger H U$ with $U = \exp(iAt)$ and $A$ is given by

$$A = \begin{pmatrix} \omega & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \omega \end{pmatrix}.$$

Then, the transformation operators are

$$U = \begin{pmatrix} e^{i\omega t} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & e^{i\omega t} \end{pmatrix}, \quad U^\dagger = \begin{pmatrix} e^{-i\omega t} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & e^{-i\omega t} \end{pmatrix},$$

and the spin operators transform according to
where in the last row we assigned creases, and the signal decreases substantially as the Rabi frequency in-
population transfer at the first minimum point in the exceeded. In contrast, in ref. \( \phi \)
edge, aഗmany realizations of the MW phase at the pulserising
tal technique which was used in our work namely to aver-
to predict. The contridiction arises from the experimen-
π
driving regime, and that the unity population transfer can occur also in the strong

The terms in the Hamiltonian are calculated as follows

\[
U^\dagger S_x U = \frac{1}{\sqrt{2}} \begin{bmatrix} e^{-i\omega t} & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & e^{-i\omega t} \end{bmatrix} \begin{bmatrix} 0 & 1 & 0 \\
1 & 0 & 0 \\
0 & 0 & 0 \end{bmatrix} = \frac{1}{\sqrt{2}} \begin{bmatrix} 0 & e^{-i\omega t} & 0 \\
e^{i\omega t} & 0 & e^{i\omega t} \\
0 & e^{-i\omega t} & 0 \end{bmatrix}
\]

\[
U^\dagger S_y U = \frac{1}{\sqrt{2}} \begin{bmatrix} e^{-i\omega t} & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & e^{-i\omega t} \end{bmatrix} \begin{bmatrix} 0 & -i & 0 \\
i & 0 & -i \\
i & 0 & i \end{bmatrix} = \frac{1}{\sqrt{2}} \begin{bmatrix} 0 & -ie^{-i\omega t} & 0 \\
 ie^{i\omega t} & 0 & -ie^{i\omega t} \\
0 & ie^{-i\omega t} & 0 \end{bmatrix}.
\]

The terms in the Hamiltonian are calculated as follows

\[
\Omega \cos (\omega t) U^\dagger S_x U = \Omega \frac{1}{2} \left( e^{i\omega t} + e^{-i\omega t} \right) \frac{1}{\sqrt{2}} \begin{bmatrix} 0 & e^{-i\omega t} & 0 \\
e^{i\omega t} & 0 & e^{i\omega t} \\
0 & e^{-i\omega t} & 0 \end{bmatrix} = \frac{\Omega}{2\sqrt{2}} \begin{bmatrix} 0 & 1 + e^{-2i\omega t} & 0 \\
1 + e^{2i\omega t} & 0 & 1 + e^{2i\omega t} \\
0 & 0 & 0 \end{bmatrix}
\]

\[
\cos (\omega t + \phi) U^\dagger S_y U = \Omega \frac{1}{2} \left( e^{i(\omega t+\phi)} + e^{-i(\omega t+\phi)} \right) \frac{1}{\sqrt{2}} \begin{bmatrix} 0 & -ie^{-i\omega t} & 0 \\
 ie^{i\omega t} & 0 & ie^{i\omega t} \\
0 & ie^{-i\omega t} & 0 \end{bmatrix} = \frac{\Omega}{2\sqrt{2}} \begin{bmatrix} 0 & e^{i(2\omega t+\phi)} + e^{-i(2\omega t+\phi)} \\
e^{i(2\omega t+\phi)} + e^{-i(2\omega t+\phi)} & 0 & e^{i(2\omega t+\phi)} + e^{-i(2\omega t+\phi)} \\
0 & 0 & 0 \end{bmatrix},
\]

and combine the two terms, we write the Hamiltonian in the MW rotating frame \( H'_{\text{MW}} \) :

\[
H'_{\text{MW}} = \frac{\Omega}{2\sqrt{2}} \begin{bmatrix} 0 & 1 + e^{-2i\omega t} & 0 \\
1 + e^{2i\omega t} & 0 & 1 + e^{2i\omega t} \\
0 & 0 & 0 \end{bmatrix}
\]

\[
+ \frac{\Omega}{2\sqrt{2}} \begin{bmatrix} 0 & e^{i(2\omega t+\phi)} + e^{-i(2\omega t+\phi)} \\
e^{i(2\omega t+\phi)} + e^{-i(2\omega t+\phi)} & 0 & e^{i(2\omega t+\phi)} + e^{-i(2\omega t+\phi)} \\
0 & 0 & 0 \end{bmatrix}
\]

\[
= \frac{\Omega}{\sqrt{2}} \begin{bmatrix} 0 & \varepsilon_- & 0 \\
\varepsilon_+ & 0 & \varepsilon_- \\
0 & \varepsilon_+ & 0 \end{bmatrix} + \frac{\Omega}{\sqrt{2}} \begin{bmatrix} 0 & e^{2i\omega t} & 0 \\
e^{2i\omega t} & 0 & e^{2i\omega t} \\
0 & e^{2i\omega t} & 0 \end{bmatrix}.
\]

where in the last row we assigned \( \varepsilon_+ = e^{i\phi} \left( 1 - ie^{i\phi} \right) / 2 \).

Appendix B: Global phase dependence in the linear polarization data

In the main text, Fig. 3d,e present the analysis of Rabi oscillations with linear polarized MW field: the population transfer at the first minimum point in the signal decreases substantially as the Rabi frequency increases, and the π-pulse duration (the evolution time until the first minimum) changes when the RWA is exceeded. In contrast, in ref.12 it is found that close-to-unity population transfer can occur also in the strong driving regime, and that the π-pulse duration is very hard to predict. The contridiction arises from the experimental technique which was used in our work namely to average many realizations of the MW phase at the pulse rising edge, \( \phi_g \). In[12,13], the MW phase was synchronized to the pulse edge. While our technique supresses the sensitivity to imperfections in the driving system, it occupancies a systematic reduction in the driving perofmances. It worth mentioning that there is no systematic deterioration when applying strong circular MW fields. Here we describe the global-phase dependence using numerical simulations, compared with the measured Rabi oscillations signals. Fig. 5a-c show the associated dynamics for various field strengths, and for various \( \phi_g \)-values. At \( \lambda = 0.1 \) (Fig. 5a), the influence of the counter rotating term is negligible and the dynamics is identical for any \( \phi_g \) at the appropriated rotating frame (red-dashed curves). Then, the spin rotates around a big circle on the Bloch sphere. At \( \lambda = 0.33 \) (Fig. 5b), the dynamics changes for each \( \phi_g \), but the averaged time trace still resembles harmonic oscillations (blue solid curve). This demonstrates the robustness of the averaging technique. At higher Rabi frequency \( \lambda = 1.2 \) (Fig. 5c), the counter rotating term influences the spin rotations markedly and
Figure 5: Global phase dependance in the strong driving regime. (a-c) Rabi oscillations for increasing Rabi-frequency values, $\lambda = 0.1$(a), $\lambda = 0.33$(b), $\lambda = 1.2$(c). On the left - Bloch sphere representation of the spin state. On the right - the spin’s z-component as a function of the driving duration. Red dashed curves are various realizations of $\phi_g$, and the solid blue curve is an averaged trajectory over 300 realizations. (d) Comparison between experimental values (circles, rectangles), and numerical simulations for high $\lambda$ values (1.2, and 1.4 respectively), the green curve is of Rabi oscillations at the weak driving regime and serves as guide for the eyes.

for each $\phi_g$ the dynamics is complete different; while for a given $\phi_g$ complete population transfer from $\langle \sigma_z \rangle = +1$ to $\langle \sigma_z \rangle = -1$ can occur (Fig. 5c, red thick dashed curve), for the ensemble-average of many $\phi_g$, the population transfer doesn’t exceed 60%, demonstrating the downside of this technique. When averaging many $\phi_g$ realizations, the principle minimum point of the signal, which marks the $\pi$-pulse operation, changes into multiple minima structure, as shown in Fig. 5d. The circles and rectangles are experimental values measured at $\lambda = 1.2, 1.4$, respectively. The solid lines are the results of a numerical simulation averaging 300 different $\phi_g$-values. From the Bloch sphere representation it is clear that at these values the averaged spin-state becomes mixed (red, blue curves) compared to the rotation at the weak driving regime (green curve). In Fig. 5d, we have marked the points “A” and “B” which are addressed in the main text and in Fig. 5.