Environmentally Mediated Coherent Control of a Spin Qubit in Diamond

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The coherent control of spin qubits forms the basis of many applications in quantum information processing and nanoscale sensing, imaging, and spectroscopy. Such control is conventionally achieved by direct driving of the qubit transition with a resonant global field, typically at microwave frequencies. Here we introduce an approach that relies on the resonant driving of nearby environment spins, whose localized magnetic field in turn drives the qubit when the environmental spin Rabi frequency matches the qubit resonance. This concept of environmentally mediated resonance (EMR) is explored experimentally using a qubit based on a single nitrogen-vacancy (NV) center in diamond, with nearby electronic spins serving as the environmental mediators. We demonstrate EMR driven coherent control of the NV spin state, including the observation of Rabi oscillations, free induction decay, and spin echo. This technique also provides a way to probe the nanoscale environment of spin qubits, which we illustrate by acquisition of electron spin resonance spectra from single NV centers in various settings.

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The coherent control of spin qubits is fundamental to endeavors in both quantum computing and nanoscale sensing. In quantum computing, the ability to coherently control the spin state of a target qubit within an array is essential to quantum information processing, and to harnessing the enhanced computing power of quantum algorithms [1–4]. In quantum sensing, the coherent control of a qubit spin state is required to selectively decouple the qubit from its magnetic environment, enhancing sensitivity to some target signal window [5–10]. This has led to a significant decrease in sensing volumes as compared to conventional magnetic resonance experiments [11,12], achieving detection at the single-electron-spin level [13,14], and holds promise towards atomic-resolution imaging of single biomolecules [15–19].

Coherent control of qubit spin states is typically achieved directly, by application of a global driving field resonant with a qubit transition, while the fluctuating states of spins present in the local qubit environment decohere the qubit state. The unwanted decoherence caused by these environmental spins typically limits the ability to perform complex algorithms for quantum computation, or, equivalently, limits the performance of the qubit for sensing purposes. Here we present a technique by which these typically chaotic environmental spins are appropriated as localized agents of control, allowing the coherent manipulation of a proximal qubit state. Precisely, control of the qubit is achieved by matching the Rabi frequency of directly driven environmental spins with the qubit spin-transition frequency. This environmentally mediated resonance (EMR) condition therefore classifies as a Hartmann-Hahn-like double resonance [20–24].

To demonstrate this concept experimentally, we use a qubit based on a single nitrogen-vacancy (NV) defect center in diamond, which can be optically initialized and readout under ambient conditions [25], and enlist an ensemble of nearby electron spins as environmental mediators. The resonance landscape of EMR is explored by varying driving frequency, driving strength, and external field strength, and is found to be in good agreement with a simple semiclassical model. Coherent control of the NV spin state is illustrated by performing EMR driven analogues of Rabi, free induction decay, and spin-echo experiments. Finally, applications to nanoscale spectroscopy are demonstrated, including the acquisition of a substitutional nitrogen spectrum by EMR driving. We point out that our work shares similarities with previous work by London et al. [22], which uses a driven NV qubit to address nearby nuclear spins through a Hartmann-Hahn resonance.

Principle.—The resonance landscape of EMR driving can be understood by a simple semi-classical model, formulated by double application of the Rabi formula, first applied to the environmental spins under direct driving, modelled as a single macrospin for simplicity, and second to the qubit, here the NV center, as driven by the effective field arising from the environmental spin Rabi oscillations [Fig. 1(a)]. Assuming the NV spin is initialized in the |0⟩ state, this model gives the probability of measuring the |−1⟩ state after a driving time, τ, as [26]

\[ P_{-1}(\tau) = \left( \frac{\Omega_{NV}}{\Omega_{NV}} \right)^2 \frac{1 - \cos(\Omega_{NV} \tau)}{2} \]
where $\Omega_{NV} = \sqrt{\Omega_{env}^2 + (\omega_{MW} - \omega_{env})^2}$ is the Rabi frequency of the NV, with an effective driving strength $\Omega_{env} = \gamma_e \alpha(\{r_i\})/(\Omega_{env}/\Omega_{env})^2/\sqrt{2}$. The latter is dependent on the relative amplitude of the environmental spin Rabi oscillations, $(\Omega_{env}/\Omega_{env})^2$, and the net magnetic field projection perpendicular to the NV axis due to the ensemble of environmental spins, $\alpha(\{r_i\})$, at positions $\{r_i\}$ relative to the NV [26]. The environmental spin Rabi frequency is given by $\Omega_{env} = \sqrt{\Omega_{env}^2 + (\omega_{MW} - \omega_{env})^2}$, with driving strength $\Omega_{env} = \gamma_e B_1/2$, where $\gamma_e$ is the electron gyromagnetic ratio and $B_1$ is the direct driving magnetic field amplitude. The NV $|0\rangle \leftrightarrow | -1\rangle$ spin transition frequency is denoted as $\omega_{NV}$, that of the spin-$1/2$ environmental spins as $\omega_{env}$, and the driving microwave field frequency is $\omega_{MW}$.

Equation (1) suggests an EMR matching condition when the environmental spin Rabi frequency, or, equivalently, the dressed state transition frequency, is brought into resonance with the NV transition frequency, $\omega_{NV} = \Omega_{env}$ [Fig. 1(b)], which gives

$$\omega_{NV} = \sqrt{\Omega_{env}^2 + (\omega_{MW} - \omega_{env})^2},$$

leading to maximal probability oscillation between the $|0\rangle$ and $| -1\rangle$ states of the NV spin. Note that there exists an optimal EMR driving condition when the environmental spins are driven resonantly, $\omega_{MW} = \omega_{env}$, maximizing the resulting NV Rabi frequency, $\Omega_{NV}$, within the matching condition, Eq. (2).

**Experiment.**—EMR driving of a single NV center by an ensemble of environmental electron spins is achieved using an electronic-grade diamond crystal with $^{15}$NV centers implanted 5–15 nm below the surface [26]. Free-electron spins known to exist at the diamond surface comprise the environmental spin ensemble [Fig. 1(a)] [28–32]. Because of the 2.87 GHz zero-field splitting between the $|0\rangle$ and $| -1\rangle$ NV spin states, and experimental difficulties in achieving GHz Rabi frequencies of the environmental spins, the EMR matching condition is most conveniently achieved near the ground-state level anticrossing (GSLAC), which occurs at an external field strength $B_0 = 1024$ G [Fig. 1(c)]. These experiments are performed at external field strengths giving $\omega_{env}$ in the range 0–10 MHz, and $\omega_{env}$ in the range 2860–2880 MHz, accordingly.

In this regime, the NV electronic spin-state structure is complicated by hyperfine interaction with the intrinsic nuclear spin of the NV, giving rise to multiple hyperfine shifted transitions [33,34]. To simplify the EMR resonance landscape, an external field of $B_0 \approx 1023$ G was chosen, giving a single degenerate NV transition frequency at $\omega_{NV} \approx 2.65$ MHz, as determined by directly driven magnetic resonance [Fig. 2(a)]. Here the photoluminescence (PL) is a measure of the population of the $|0\rangle$ state, and the decrease on resonance indicates driving of the $|0\rangle$ to $| -1\rangle$ transition when $\omega_{MW} = \omega_{NV}$.

To measure the EMR landscape, we use the pulse sequence illustrated in Fig. 2(b), where the microwave pulse frequency $\omega_{MW}$ is swept across the environmental spin transition frequency $\omega_{env}$ and the driving field amplitude, $\Omega_{env} = \gamma_e B_1/2$ [26], is swept across the NV transition $\omega_{NV}$. The microwave pulse duration $\tau$ is fixed to maximize PL contrast at the optimal EMR driving condition, and a single laser pulse is used for optical readout and repumping of the NV spin state. Figures 2(c) and 2(d) show EMR PL maps as a function of $\omega_{MW}$ and $\Omega_{env}$ as measured in experiment and predicted by Eq. (1), respectively. The dashed lines show the EMR matching condition given by Eq. (2), centered about the optimal driving condition where $\omega_{MW} = \omega_{env} = 2868$ MHz, and $\Omega_{env} = \omega_{NV} = 2.65$ MHz. The resonant branches emanating from this point arise from the ability to recover the matching condition $\Omega_{env} = \omega_{NV}$, when the environmental spins are driven off resonance, $\omega_{MW} \neq \omega_{env}$, by reducing the driving strength, $\Omega_{env}$. The experimental data are found to be in good overall agreement with the theoretical model, as indicated by the line cuts presented in Figs. 2(e) and 2(f). The broadening of the experimental map in driving strength as compared to the theoretical plot is attributed to magnet drift throughout the acquisition time [10 h for Fig. 2(c)].

In general there are two $^{15}$NV hyperfine transitions about the GSLAC, which overlap at $B_0 = 1023$ G [34]. Repeating the previous measurement at various external field strengths across the GSLAC reveals this hyperfine structure as multiple resonance features in the EMR.
These EMR features match, in terms of $\bar{\Omega}$, the hyperfine transitions resolved by direct driving of the NV (right-hand side in Fig. 3), with matching PL contrasts. This demonstrates the ability to selectively drive NV hyperfine transitions with EMR driving, by virtue of the relatively low power of the local driving fields involved. In addition, the center of these resonance features shifts in driving frequency with increasing external field strength, in accordance with the Zeeman splitting of the free-electron spin states, $\omega_{\text{env}} = \gamma_e B_0$ [see Fig. 1(c)].

These measurements, which utilized a fixed driving pulse duration maximizing PL contrast, demonstrate the ability to induce spin transitions of a target NV center by EMR driving. We now probe the EMR driving dynamics by time resolved measurements, allowing the coherence of the observed control to be assessed. Utilizing the optimal driving parameters $\omega_{\text{MW}} = \omega_{\text{env}}$ and $\bar{\Omega}_{\text{env}} = \omega_{\text{NV}}$ identified in Fig. 2(c) at the $^{13}$NV hyperfine crossing, an EMR driven Rabi curve on the NV was measured by varying the driving pulse duration [Fig. 4(a)]. An oscillation with a period of 3 $\mu$s is observed, demonstrating coherent control of the NV spin state. The corresponding Rabi frequency, $\Omega_{\text{NV}} = 3.3$ MHz, is intrinsically linked to the spatial distribution of environmental spins, offering a pathway towards spatial mapping of such spins with nanoscale resolution. The rapidly decaying envelope of the Rabi curve arises as a consequence of the random initial spin state of the environmental spin ensemble, such that the resulting curve is an average across a distribution of effective driving strengths of the environmental spin field [26].

We note that EMR Rabi driving can be achieved for any pair of driving parameters $(\omega_{\text{MW}}, \bar{\Omega}_{\text{env}})$ satisfying the EMR matching condition depicted by the dashed line in Fig. 2(c). This is illustrated in Fig. 4(b), showing EMR Rabi curves as a function of $\bar{\Omega}_{\text{env}}$, with the driving frequency $\omega_{\text{MW}}$ chosen such that the EMR matching condition is satisfied where
oscillation at the detuning frequency \[35\]. The spin-echo directly driven Ramsey measurements, which exhibit a time of the sequence \[26\]. This feature is in contrast with microwave pulse sequence as inset.

EMR spin-echo curve using pulse durations identified in (a), with \(2.65 \text{ MHz}\) as determined by fit (solid line). (d) Optimally driven shows a characteristic oscillation at the NV transition frequency, driving parameters and identified satisfied. (c) EMR driven Ramsey measurement using the optimal driving pulse sequence as inset.

Contrast \[\text{lower half Fig. 4(b)}\]. Decreasing the driving \(\omega_{\text{MW}}\) for a given environmental spin ensemble \[26\], and can, therefore, be improved by using an \(^{14}\text{N}\) center, for which \(\omega_{\text{NV}}\) can typically be reduced to 100 kHz \[34\].

Applications of EMR to spectroscopy are made particularly attractive due to its experimental simplicity as compared to competing techniques, such as double electron-electron resonance \[26\], which requires pulsed driving of the qubit probe and environmental spin species in parallel \[28,40\], and \(T_1\)-based spectroscopy, which requires fine scanning of the aligned magnetic field possible. When \(\Omega_{\text{env}} > \omega_{\text{NV}}\), the driving frequency is fixed at \(\omega_{\text{MW}} = \omega_{\text{env}}\), as the EMR matching condition cannot be recovered in this regime, resulting in a sharp decrease in PL contrast [lower half Fig. 4(b)]. Decreasing the driving strength below the optimal condition \(\Omega_{\text{env}} < \omega_{\text{NV}}\) preserves the contrast, but gives a longer Rabi period according to the factor \((\Omega_{\text{env}}/\omega_{\text{NV}})^2\) [upper half Fig. 4(b)].

The coherent control demonstrated in Figs. 4(a) and 4(b) suggests the feasibility of using EMR to drive pulsed quantum control schemes fundamental to quantum information and quantum sensing protocols. Identifying effective \(\pi/2\)- and \(\pi\)-pulse durations from the optimally driven EMR Rabi curve at 650 and 1300 ns, respectively, Ramsey and spin-echo measurements were performed. The free induction decay curve as measured by the EMR driven Ramsey sequence [Fig. 4(c)] shows an oscillation at approximately 2.65 MHz, the NV transition frequency. This oscillation arises from the phase accumulation of the NV spin state relative to the effective driving field of the environmental spin ensemble, whose phase is effectively frozen during the free evolution time \[26\]. An analytic treatment in the macrospin approximation of the NV state evolution under this driving scheme reveals this oscillation, giving the probability of measuring the \(|0\rangle\) state as \(P_0(\tau) = [1 - \cos(\omega_{\text{NV}} \tau)]/2\), where \(\tau\) is the free evolution time of the sequence \[26\]. This feature is in contrast with directly driven Ramsey measurements, which exhibit an oscillation at the detuning frequency \[35\]. The spin-echo sequence, by design, filters out effects from quasistatic dephasing processes \[26\]. Consequently, the EMR driven spin-echo curve [Fig. 4(d)] shows revivals at a frequency of 1.1 MHz, corresponding to the Larmor precession of the surrounding bath of \(^{13}\text{C}\) nuclear spins \[36\]. In addition, theoretical analysis shows that the decay of the Ramsey and spin-echo measurements are dominated by the decoherence of the NV, with characteristic time scales \(T_1\) and \(T_2\), respectively, and the randomized initial states and decoherence of the environmental spins result primarily in a reduced contrast \[26\].

As a final experiment, we illustrate the applicability of EMR to spectroscopy by acquiring an electron spin resonance spectrum of a nontrivial spin species, namely, substitutional nitrogen (P1) centers internal to a nitrogen rich host diamond [Fig. 5(a)]. A representative P1 spectrum acquired by EMR driving is given in Fig. 5(b), revealing the characteristic five-peak structure of the center due to the on- and off-axis parallel hyperfine interaction between the spin-\(^{14}\text{N}\) nuclear spin and spin-\(1/2\) electron spin of the center \[37–39\]. We note that the resonance line width, which sets the spectral resolution of the technique, is governed by \(\omega_{\text{NV}}\) for a given environmental spin ensemble \[26\], and can, therefore, be improved by using an \(^{14}\text{N}\) center, for which \(\omega_{\text{NV}}\) can typically be reduced to 100 kHz \[34\].

Applications of EMR to spectroscopy are made particularly attractive due to its experimental simplicity as compared to competing techniques, such as double electron-electron resonance \[26\], which requires pulsed driving of the qubit probe and environmental spin species in parallel \[28,40\], and \(T_1\)-based spectroscopy, which requires fine scanning of the aligned magnetic field.

FIG. 4. (a) Optimally driven EMR Rabi curve with \(\omega_{\text{MW}} = \omega_{\text{env}}\) and \(\Omega_{\text{env}} = \omega_{\text{NV}}\) at the \(^{15}\text{NV}\) hyperfine crossing, \(\omega_{\text{NV}} = 2.65 \text{ MHz}\), with microwave pulse scheme as inset. Effective \(\pi/2\)- and \(\pi\)-pulse times are identified at 650 and 1300 ns, respectively. (b) EMR driven Rabi PL map as a function of driving pulse duration and driving strength. Optimally driven Rabi curve using parameters identical to (a) is highlighted.

FIG. 5. (a) EMR schematic for driving of substitutional nitrogen (P1) center electron spins (red) in bulk diamond. P1 center nuclear spins (gray) lead to hyperfine splitting of the electron spin transition. (b) Characteristic P1 center spectrum acquired by EMR spectroscopy at \(B_0 \approx 1024 \text{ G}\). The FWHM of central resonance peak is approximately 5 MHz. (b) Continuous wave (cw) and pulsed EMR spectra of near surface free-electron species, with control schemes given as insets. Offset between the spectra is due to a variation in the external field strength used for each measurement.
The EMR protocol can be further simplified by implementing a continuous wave (cw) optical and microwave excitation scheme, achieving similar results as compared to the pulsed scheme [Fig. 5(b)]. The reduced fluorescence contrast of the cw scheme is ascribed to the continuous optical repumping of the NV spin state [42].

In this Letter we have introduced a technique by which the coherent driving of a qubit is achieved by using nearby environmental spins as agents of control. This concept has been realized in experiments using a single NV center in diamond, driven by an ensemble of electron spins, both at the diamond surface and in the bulk. The parameter space of this technique has been explored and compared to the simple semiclassical model developed, showing good agreement. Applications to spectroscopy have been demonstrated by acquisition of a characteristic substitutional nitrogen center spectrum, and are made attractive by the experimental simplicity of the technique. Finally, the highly localized driving fields utilized by EMR provide an avenue by which target qubits within an array can be selectively addressed, especially in conjunction with environmental spin engineering.

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