Supporting Online Material for

Coherent Dynamics of a Single Spin Interacting with an Adjustable Spin Bath

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I. EXPERIMENTAL METHODS

A. Measurement setup

The NV center spins are imaged, initialized and read out using a confocal microscope at room temperature. The magnetic field is aligned along the [111] axis of the diamond sample. The NV centers studied in this work are formed during growth in a type Ib single-crystal diamond from Sumitomo Electric. This diamond contains nitrogen (N) impurities with a density of $10^{19} - 10^{20}$ cm$^{-3}$. The dipolar interaction strength between electron spins is therefore of order MHz.

Single NV centers are identified by observing antibunching in photon correlation measurements (demonstrating that it is a single emitter) and measurement of the spin splitting at zero magnetic field (showing it is an NV center). The NV centers studied here were selected to have their symmetry axes parallel to the magnetic field, so that mixing of electronic spin levels is minimized. Details about the optics part of the setup, the identification of single NV centers and about the magnetic field alignment can be found in Ref. [1].

We use magnetic resonance to coherently manipulate the electron spin state. An oscillating radiofrequency (RF) current is sent through a thin gold wire (10 micrometer diameter) which generates an oscillating RF magnetic field. The wire is positioned to within about 20 micrometer of the NV center under study. Multiple synchronized outputs of a pattern generator (HP 81130A) are used to control the laser via an acoustic-optical modulator (Isomet 1250C), to gate the photon counters, and to trigger an arbitrary wave-form generator (Tektronix AWG520). The arbitrary waveform generator in turn chops a continuous-wave RF signal (Agilent E8257C) through an RF switch (Minicircuits ZA-SWA-2-50DR), leading to the desired sequence of RF bursts. Risetime of the RF pulses after the switch is about 1 nanosecond. In order to obtain high driving powers the RF signal is amplified with up to 40 dB (Amplifier Research 25S1G4A).

B. Spin manipulation sequences

The pulse duration needed to achieve a 90-degree rotation ($\pi/2$ pulse) or a 180-degree rotation ($\pi$ pulse) of the spin is calibrated from coherently driven spin oscillations. All pulse sequences start by initializing into $|m_S = 0\rangle$. At $B = 0$ G, all three spin sublevels
are manipulated. A $\pi/2$ pulse creates the spin superposition state $\frac{1}{\sqrt{2}}(|m_S = -1\rangle + |m_S = +1\rangle)$ (up to a global phase). In the Ramsey experiments, dephasing of this state is measured by applying a second $\pi/2$ pulse after a variable free evolution time. In case no phase difference is acquired, this second pulse brings the system back to the initial state $|m_S = 0\rangle$. At $B = 740$ G, we only manipulate the spin sublevels $|m_S = 0\rangle$ and $|m_S = -1\rangle$ (the state $|m_S = +1\rangle$ is far away in energy and does not contribute to the dynamics). A $\pi/2$ pulse on this effective two-level system (effective spin 1/2) creates the spin superposition state $\frac{1}{\sqrt{2}}(|m_S = -1\rangle + |m_S = 0\rangle)$ (up to a global phase). In the Ramsey experiments, the second $\pi/2$ pulse now creates the state $|m_S = -1\rangle$ in case of no dephasing. As a consequence, the Ramsey curves at $B = 0$ G and at $B = 740$ G have opposite initial values in our experiments (see Fig. 2A).

To measure the coherence time $T_2$ a $\pi$ pulse is inserted in between the two $\pi/2$ pulses, see Fig. S1A. Figure S1B shows measurements on NV31 at $B = 0$ G where the free evolution time before the $\pi$ pulse, $\tau_1$, is fixed, and the free evolution time after the $\pi$ pulse, $\tau_2$, is varied. A clear echo signal is obtained whenever $\tau_1 = \tau_2$, demonstrating that part of the dephasing can be reversed. Note that both at $B = 0$ G and at $B = 740$ G, the echo sequence ideally brings the system back to the initial state $|m_S = 0\rangle$ (up to a phase factor), since the total rotation is over an angle of $2\pi$. As expected, the observed oscillations as a function of $\tau_2$ are similar to those obtained from the Ramsey experiments (see Figure 2A). By setting $\tau_1 = \tau_2 = \tau$ and varying the total free evolution time $2\tau$, a spin echo measurement is performed (see Figure S1C).

II. THEORETICAL DESCRIPTION OF THE SYSTEM

We consider decoherence of a single NV center in diamond by a bath of the electron spins of the nitrogen atoms (P1 centers, according to classification of Ref. [2]). The NV center is treated as a localized spin $S_0 = 1$, possessing a single-axis anisotropy $H_0^g = D(S_0^g)^2$, where $D = 2.87$ GHz is the splitting between the levels $|m_S = 0\rangle$ and $|m_S = \pm 1\rangle$ (here $m_S$ denotes the eigenstates of $S_0^g$) at zero external magnetic field. The anisotropy $z$-axis is directed along the [111] direction. The quantity measured in experiments is $p_0(t)$: the time-dependent population probability of the state $|m_S = 0\rangle$. The initial state of the NV center is $|m_S = 0\rangle$, while the state of the bath is maximally mixed: the density matrix of the
bath is proportional to the identity matrix, i.e. all states of the bath are equally probable.

The nitrogen P1 centers are treated as localized spins $S_k = 1/2$ ($k = 1, \ldots, N$). For given experimental circumstances (large average distance between the NV center and the nitrogens), the most dominant interaction between the NV center and the nitrogens (the system-bath interaction $H_{SB}$) is the dipolar interaction. Moreover, the dipolar interaction between different bath spins is also relevant, since the typical distance between different nitrogens is the same as the typical distance between NV and a nitrogen. Finally, it is important to take into account the hyperfine interactions on the NV site and on the P1 sites, since the electron spins at both NV and P1 sites interact with the nuclear spin of $^{14}$N.

Decoherence of a central spin by a spin bath is a very complex many-body non-equilibrium quantum phenomenon, and it is important in many areas of physics, from magnetic resonance to quantum information processing. The proper and adequate description of the spin-bath decoherence of a central spin is a rather old problem, and many theoretical approaches of different degree of sophistication have been developed for different interesting solid-state systems, starting from early days of NMR/ESR theory (e.g., [3, 4]) till very recently (see, e.g., Reference 3-10 of the main text). All these theories involve a trade-off between quantitative rigour and qualitative understanding. Some of these theories use simplified phenomenological treatments and involve certain assumptions about the system’s behavior, but allow all calculations to be performed analytically, and give a very clear (although crude) qualitative picture of the underlying physics. Other theories use more detailed numerical simulations, with fewer (or no) assumptions, and gain quantitative rigour at the expense of being much less clear. In the present work, we use two theoretical approaches, located at the opposite ends of this dilemma. First, we use a simple phenomenological model which allows for a completely analytical treatment, and provides qualitative understanding of experiments. Second, we use direct numerical simulations, starting straight from the relevant microscopic Hamiltonians; this simulations give much less qualitative insight, but involve absolutely no assumption about the behaviour of the system. The agreement between both approaches and the experimental results allows us to achieve both goals: to gain qualitative insights into the dynamics of decoherence of an NV center, and, at the same time, to make sure that our qualitative physical picture is correct.

Below, in Sections III–IV, we explain in detail the microscopic structure and derivation of the Hamiltonians for the NV and P1 centers. Explanation of a simplified analytical model
for the bath of P1 centers, and the analytical results are given in Sec. V. The details of numerical simulations are given in Sec. VI.

III. HAMILTONIAN OF A SINGLE NV CENTER

Along with the single-axis anisotropy mentioned above, another important interaction is the hyperfine coupling on the NV site, i.e. the coupling between the electron spin $S_0 = 1$ and the nuclear spin $^{14}$N of the NV center (denoted below as $I_0$). The relevant hyperfine Hamiltonian is deduced from the experimental measurements in Refs. [7, 8]:

$$H_{hf}^0 = A_0 S_0^z I_0^z + A_1 (S_0^x I_0^x + S_0^y I_0^y) - P_0 (I_0^z)^2,$$

where $A_0 = 2.3$ MHz, $A_1 = 2.1$ MHz, and $P_0 = -5.1$ MHz. Also, external magnetic fields can be applied to the system: (1) a static magnetic field $B$ along the z-axis ([111] direction), and/or (2) an oscillating field $H_R$ with frequency $\omega$ can be applied to the NV center along the $x$-axis, either in order to study Rabi oscillations, or to implement the $\pi$- and $\pi/2$-pulses in the Ramsey and Hahn spin echo experiments. Thus, the relevant part of the NV center Hamiltonian is

$$H_S = D (S_0^z)^2 + A_0 S_0^z I_0^z + A_1 (S_0^x I_0^x + S_0^y I_0^y) - P_0 (I_0^z)^2 + g_0 \mu_B B S_0^z + g_0 \mu_B H_R S_0^x \cos \omega t,$$

where $g_0 = 2$ is the Landé factor of the NV center, and $\mu_B$ is Bohr’s magneton. This Hamiltonian is complex, and inconvenient for numerical simulations, as it contains large energy scales: $D$ and, in case of large magnetic field, $g_0 \mu_B B$. Significant simplification can be achieved by going to the interaction representation, which excludes the large terms from the Hamiltonian (1), and treating the remaining terms perturbatively.

Below, we perform this simplification for two experimentally relevant situations: zero magnetic field $B = 0$, and large magnetic field $B = 740$ G. For the readers familiar with the NMR/ESR theory, the essence of the derivation below can be formulated concisely: we perform the standard procedure of rotating-frame transformation and omission of the non-secular (explicitly time-dependent) terms [10].
A. Zero magnetic field, \( B = 0 \)

At zero magnetic field, we eliminate the first term by applying a unitary transformation 
\[ U_1 = \exp \left[ -iD(S_0^z)^2t \right] = 1 + (S_0^z)^2[\exp(-iDt) - 1] \] to the Hamiltonian (1), treating the term \( D(S_0^z)^2 \) as a zero-order Hamiltonian, and everything else as a perturbation. In this way, the terms containing the operators \( S_0^x \) and \( S_0^y \) become time-dependent, oscillating with the frequency \( D \). Due to these oscillations, for instance, the third term in (1) is averaged out: its time-average value is zero, so it gives no contribution in the first-order approximation. In the second order, the effect of this term is twofold. Firstly, it mixes the state \( |m_S = 0\rangle \) with \( |m_S = \pm 1\rangle \) and slightly renormalizes their energies. These effects are small \( (A_1/D \sim 10^{-3}) \), and can be neglected: e.g., the energy corrections are of order of \( A_1^2/D \sim 1.5 \) kHz, and they can be visible only at timescales of few milliseconds, much larger than the relevant experimental timescales (few microseconds). Secondly, the term \( A_1(S_0^x I_0^x + S_0^y I_0^y) \) can strongly mix the states \( |m_S = +1\rangle \) and \( |m_S = -1\rangle \), since they are degenerate in zeroth order. However, this effect is also too weak, and any external field larger than \( A_1^2/D \sim 10^{-3} \) G already destroys it (this field can come, for instance, from interaction with the bath, which is of order of 0.1 G). Finally, we note that since this term does not affect the electron spin \( S_0 \), it also does not affect the nuclear spin \( I_0 \), because the flip-flop processes involve both \( S_0 \) and \( I_0 \). The conclusion is that the third term in (1) can be safely dropped. The nuclear quadrupolar term \( P(I_0^z)^2 \) then becomes just a constant, and can also be ignored.

However, the last term in the Hamiltonian (1) requires more care: it is already explicitly time-dependent, and after the unitary transformation \( U_1 \), its dependence on time has the form of either \( \exp(iDt) \cos \omega t \) or \( \exp(-iDt) \cos \omega t \), depending on the position in the \( S_0^z \) matrix. In zero-magnetic field experiments, the frequency of the oscillating field is \( \omega = D \), so that the terms \( \exp(\pm iDt) \cos \omega t \) contain both non-oscillating part \( 1/2 \) and the part oscillating with the frequency \( 2D \). The oscillating part has negligible effect: all considerations of the previous paragraph can be applied here with minor modifications. Thus, we retain only the non-oscillating (secular) part, and the Hamiltonian of the NV center then becomes

\[ H_S = A_0 S_0^z I_0^z + (1/2)g_0 \mu_B H_R S_0^z. \]  

(2)

Note that the last term, which describes the action of the oscillating driving field, excites both transitions \( |m_S = 0\rangle \leftrightarrow |m_S = 1\rangle \) and \( |m_S = 0\rangle \leftrightarrow |m_S = -1\rangle \).
Finally, we note that in this work, we do not excite the nuclear spin of the NV center, and the intrinsic relaxation time (due to e.g. spin-phonon coupling) of the spin $I_0$ is of order of milliseconds. Thus, for a single experimental run, the nuclear spin is frozen, and $I_0^z$ is the constant of motion. Then, the nuclear spin $I_0$ just creates an additional static magnetic field along the $z$-axis (equal to $\pm A_0$ or zero, depending on $m_I$, where $m_I = 0, \pm 1$ denotes the eigenstates of $I_0^z$). However, between different experimental runs, the nuclear spin has enough time to switch to a state with another $m_I$. Thus, in order to calculate the experimentally observable population $p_0(t)$ of the state $m_S = 0$, we need to perform calculations for different $m_I$, and average the result.

B. Large magnetic field, $B = 740$ G

If the magnetic field $B$ is large, then the transitions $|m_S = 0\rangle \leftrightarrow |m_S = 1\rangle$ and $|m_S = 0\rangle \leftrightarrow |m_S = -1\rangle$ have very different frequencies: $D + g_0 \mu_B B$ for the first transition, and $D - g_0 \mu_B B$ for the second one. In experiments, the driving field frequency $\omega$ is set to excite only the second transition, $\omega = D - g_0 \mu_B B$, so that the state $|m_S = +1\rangle$ is not affected at all. In this case, the NV center can be described in terms of a fictitious spin $s_0 = 1/2$, with the state $|\uparrow\rangle$ of $s_0$ corresponding to $|m_S = 0\rangle$, and the state $|\downarrow\rangle$ of $s_0$ corresponding to $|m_S = -1\rangle$. By projecting the full Hamiltonian (1) onto the subspace spanned by the levels $|m_S = 0\rangle$ and $|m_S = -1\rangle$, an effective Hamiltonian for the spin $s_0$ is obtained:

$$H_S = A_0 s_0^z I_0^z + \frac{1}{\sqrt{2}} g_0 \mu_B H_R s_0^x.$$  

(3)

where the non-secular terms are excluded in the same way as in Sec. III A (all considerations of that section are applicable here with minor modifications).

IV. HAMILTONIAN OF THE BATH, AND COUPLING OF AN NV CENTER TO THE BATH

The bath is made of nitrogen atoms (P1 centers [2]), each having an unpaired electron with the spin $S_k = 1/2$ and a $^{14}$N nuclear spin $I_k = 1$. The hyperfine coupling between $S_k$ and $I_k$ is rather strong (around 100 MHz), and should be taken into account.
A. Hamiltonian of a single P1 center

In order to describe the role of the hyperfine coupling, we need to consider the microscopic structure of a P1 center. At every P1 center, the unpaired electron is shared by the nitrogen atom and the neighboring carbon atom. For every nitrogen, there are four equivalent neighboring carbons, so the electron can be delocalized along one of four axes: [111], [111], [111], and [111]. Correspondingly, there are four types of P1 centers. For a given P1 center, let us introduce a local coordinate system $(X, Y, Z)$, with the $Z$-axis coinciding with the delocalization axis of this P1 centers. For one type of center, with the delocalization axis [111], the local coordinates $(X, Y, Z)$ coincide with the laboratory coordinates $(x, y, z)$. For three other types, $(X, Y, Z)$ differ from $(x, y, z)$ by a $90^\circ$- or a $180^\circ$-rotation around the $c$-axis of the crystal. In the local coordinate frame, the hyperfine Hamiltonian of a single P1 center is of single-axis type: [2, 9]

\[
H_L = A_Z I_k^Z S_k^Z + A_X (I_k^X S_k^X + I_k^Y S_k^Y) - P (I_k^Z)^2
\]

where $(I_k^X, I_k^Y, I_k^Z)$ are the operators of the $k$-th nuclear spin in the local coordinate frame, $(S_k^X, S_k^Y, S_k^Z)$ are the operators of the $k$-th nitrogen electron spin in the local coordinate frame, and the hyperfine and quadrupole parameters are known [2, 9]: $A_Z = 114$ MHz, $A_X = 81.3$ MHz, and $P = -4$ MHz. Note that the electron spin $S_k = 1/2$ of a P1 center has no anisotropy, so that the simplifications of Sec. III A can not be used here.

It is important to note that the delocalization axis of a given P1 center can change in time, switching from one possible direction to another. At room temperatures, the time between such switches is much longer than the characteristic time of a single experimental run. On the other hand, during the whole experiment (which includes $10^5$ single runs), the delocalization axis of a given nitrogen can switch several times. Thus, in calculations, we should consider the axis direction as static, but, in the end, average over all possible directions of the delocalization axis.

The Hamiltonian (4) has three doubly degenerate eigenenergies: $E_0 = 0$ (corresponding eigenfunctions will be denoted as $|\phi_0, +\rangle$ and $|\phi_0, -\rangle$), $E_1 = 130$ MHz (with eigenfunctions $|\phi_1, +\rangle$ and $|\phi_1, -\rangle$), and $E_2 = 149$ MHz (eigenfunctions $|\phi_2, +\rangle$ and $|\phi_2, -\rangle$). Note that the energy difference between these doubly degenerate subspaces is much larger than dipolar coupling to the NV center and to other P1 centers; this fact will be used below.
The eigenfunctions $|\phi_F, \pm\rangle$ (where $F = 0, 1, 2$) represent the entangled states of the electron and the nuclear spin of a P1 center. For instance, for a type-1 center, these functions can be expressed in the basis $|S_k^z; I_k^z\rangle$ as follows:

$$|\phi_{0,-}\rangle = -p|1/2;0\rangle + q|-1/2;1\rangle, \quad |\phi_{0,+}\rangle = -q|1/2;-1\rangle + p|-1/2;0\rangle;$$

$$|\phi_{1,-}\rangle = p|1/2;-1\rangle + q|-1/2;0\rangle, \quad |\phi_{1,+}\rangle = -q|1/2;0\rangle - p|-1/2;1\rangle;$$

$$|\phi_{2,-}\rangle = |-1/2;-1\rangle, \quad |\phi_{2,+}\rangle = |1/2;1\rangle;$$

where $p = 0.515$ and $q = 0.857$.

At large magnetic field, the Zeeman term $g\mu_B B S_k^z$ has to be added to the Hamiltonian (4), where $g = 2$ is the Landé factor of the electron spin of the P1 center; note that the field is directed along the $z$-axis of laboratory frame, not $Z$-axis of the local frame. Due to large field $B = 740$ G, the Hamiltonian can be simplified using the arguments similar to those of Sec. IIIA; the resulting Hamiltonian is

$$H_L = g\mu_B B S_k^z + A_1 S_k^z I_k^z$$

where $A_1 = 114$ MHz for the P1 centers of the type 1 (with the delocalization axis [111]), and $A_1 = 86$ MHz for the P1 centers of the other types (with the delocalization axis directed along $[\bar{1}11], [1\bar{1}1], \text{or } [11\bar{1}]$).

**B. Coupling between P1 centers and NV center**

The coupling between the NV center and the nitrogen atoms is of dipolar origin. Assuming that the NV center is located at the origin of the laboratory frame, it is:

$$H_{SB} = \sum_k \frac{g_0 g \mu_B^2}{r_k^3} [S_0 S_k - 3(S_0 n_k)(S_k n_k)]$$

where $r_k$ is the radius-vector of the $k$-th nitrogen atom, $r_k = |r_k|$, and $n_k = r_k/r_k$. Due to strong anisotropy of the NV center, this form can be simplified using the reasoning of Sec. IIIA.
1. Zero magnetic field

For the case of zero magnetic field, applying the unitary transformation $U_1$ (see Sec. III A), we obtain:

$$H_{SB} = \sum_k a_k S_0^z \left[ S_k^z - 3n_k^z (S_k^0 n_k) \right],$$

(6)

where $a_k = g_0 g \mu_B^2 / r_k^3$. However, simplicity of this expression is illusory: the local Hamiltonian of the P1 center (4) is rather complex.

To simplify matters, we note that the typical value of $a_k$ is of order of 0.5 MHz or smaller, so that this coupling Hamiltonian can be treated as a perturbation, with $H_L$ of Eq. 4 being the zeroth-order Hamiltonian. Thus, we can take into account only those matrix elements of $H_{SB}$ which connect the states of $H_L$ with equal energies, i.e. only the elements $\langle \phi_{F,\pm} | H_{SB} | \phi_{F,\pm} \rangle$ where $F = 0$, 1, or 2, and neglect all other elements. In this way, instead of complex 6-level Hilbert space of a P1 center, we have three uncoupled 2-level subspaces (indexed by $F = 0$, 1, 2). and each 2-level subspace can be described as a pseudo-spin-1/2 system, with the pseudo-spin operators $S_{k,\alpha}^\alpha$ ($\alpha = x, y, z$), where the state $| \phi_{F,\pm} \rangle$ corresponds to the state $| \uparrow \rangle$ of the pseudo-spin and the state $| \phi_{F,-} \rangle$ corresponds to the state $| \downarrow \rangle$. For instance, for a P1 center of type 1, up to possible unitary transformation, the operators $S_{k,\alpha}^{x,y,z}$ of the P1 center within these subspaces have the following nonzero elements:

$$\langle \phi_{F,+} | S_k^\alpha | \phi_{F,+} \rangle = -\langle \phi_{F,-} | S_k^\alpha | \phi_{F,-} \rangle = r_z$$

$$\langle \phi_{F,+} | S_k^\alpha | \phi_{F,-} \rangle = \langle \phi_{F,-} | S_k^\alpha | \phi_{F,+} \rangle = r_x$$

$$\langle \phi_{F,+} | S_k^\alpha | \phi_{F,-} \rangle = -\langle \phi_{F,-} | S_k^\alpha | \phi_{F,+} \rangle = r_y$$

where $r_x = 0.133$, $r_y = i \cdot 0.133$, $r_z = 0.234$ for $F = 0$; $r_x = 0.367$, $r_y = i \cdot 0.367$, $r_z = 0.234$ for $F = 1$; and $r_x = r_y = 0$, $r_z = 0.5$ for $F = 2$. Thus, for the type-1 P1 center with fixed $F$, we can simply replace $S_k^\alpha$ by $2r_\alpha S_k^{\alpha}$ ($\alpha = x, y, z$), where the operators $S_k^{\alpha}$ are unitarily equivalent to $S_k^\alpha$. The matrix elements for other types of P1 centers (with other delocalization axes) can be obtained by appropriate rotations around the c-axis of the crystal.

Since the pseudo-spin operators $S_k^\alpha$ ($\alpha = x, y, z$) are related to the spin operators $S_k^\alpha$ ($\alpha = x, y, z$) via simple unitary transformation, the Hamiltonian of interaction between the NV center and a nitrogen atom can be written as

$$H_{SB} = \sum_{k,\alpha} a_{k,\alpha}' S_0^z S_k^{\alpha}$$
By an appropriate unitary transform, this Hamiltonian can be rewritten in the simple form

$$H_{SB} = \sum_{k} \tilde{a}_k S_{0z}' \tilde{S}_{z}'_k,$$

(7)

where $\tilde{S}_{z}'_k$ is unitarily equivalent to the "original" spin operator $S_{z}'_k$, and $\tilde{a}_k = \sqrt{(a_{k,x}')^2 + (a_{k,y}')^2 + (a_{k,z}')^2}$ is proportional to $a_k$ of the "original" Hamiltonian (6); the proportionality coefficient depends on $n_k$, value of $F$, and type of the P1 center (direction of its delocalization axis).

2. Large magnetic field

For large magnetic field, the coupling Hamiltonian is simplified easily. Neither of the spins $S_0$ or $S_k$ can be flipped due to dipolar interactions, since the mismatch in energies between different states of $S_0$ or $S_k$ is very large. Formally, we take the Zeeman term (for $S_k$) and the Zeeman-plus-anisotropy term (for $S_0$) as a zeroth-order Hamiltonian, and omit the non-secular terms from the coupling Hamiltonian (6). In terms of the pseudo-spin $s_0 = 1/2$ (see Sec. III B), we obtain:

$$H_{SB} = \sum_{k} a_k [1 - 3(n_k^{z})^2] (s_0^z - 1/2) S_{z}'_k = \sum_{k} c_k s_0^z S_{z}'_k - (1/2) \sum_{k} c_k S_{z}'_k,$$

(8)

where $a_k = g_0 g \mu_B^2 / r_{jk}^3$, and $c_k = a_k [1 - 3(n_k^{z})^2]$. Note that the last term renormalizes the internal Hamiltonian of the bath.

C. Dipolar coupling between different P1 centers

The coupling between different P1 centers is important since it gives rise to the internal dynamics of the bath. The Hamiltonian of a pair of dipolarly coupled nitrogen atoms is

$$H_{jk} = H_{L,j} + H_{L,k} + H_{dip,jk},$$

(9)

$$H_{dip,jk} = g^2 \mu_B^2 \frac{1}{r_{jk}^3} [S_j S_k - 3(S_j n_{jk}) (S_k n_{jk})],$$

(10)

where $S_j$ is the electron spin of the $j$-th nitrogen, $S_k$ is the electron spin of the $k$-th nitrogen, and $r_{jk}$ is the radius-vector connecting the two nitrogens ($r_{jk} = |r_{jk}|$, and $n_{jk} = r_{jk}/r_{jk}$). The local Hamiltonians $H_{L,j}$ and $H_{L,k}$ have the form (4) or (5), depending on magnetic field; note that the local coordinate frames $(X,Y,Z)$ for the centers $j$ and $k$ may be different,
depending on how their delocalization axes are oriented with respect to the lab frame. The characteristic value of such an intra-bath coupling is of the same order as the coupling between the NV center and the P1 centers (i.e., of order of 0.5 MHz).

At zero magnetic field, the dipolar interaction leads to both single-spin flips and pairwise flip-flops. E.g., if \( k \)-th spin is in the state \( |\phi_{F,+}\rangle \) with \( F = 1 \) or \( F = 2 \), the effect of the term \( S_j^z S_k^x \) in Eq. 9 is to change the \( k \)-th spin state from \( |\phi_{F,+}\rangle \) to \( |\phi_{F,-}\rangle \), since such a transition does not change the energy and has a nonzero matrix element. In so doing, the state of the \( j \)-th spin may remain unchanged: e.g., if the \( j \)-th spin belong to a type-1 nitrogen center, then the action of \( S_j^z \) does not lead to a flip of \( j \)-th spin. On the other hand, if the \( k \)-th spin is in the state \( |\phi_{3,+}\rangle \), the only way for it to flip is to make a transition to another state, e.g. with \( F = 1 \) (or \( F = 2 \)). Such a transition changes the energy of the \( k \)-th spin; the energy conservation then requires that the \( j \)-th spin is in the state with \( F = 1 \) (or \( F = 2 \)). Thus, majority of states of the P1 centers may flip very fast, while others preserve their state for long time, being pretty much static.

At large magnetic field, the situation changes. The single-spin flips are prohibited: they do not conserve energy. But even two-spin flip-flop processes are strongly suppressed. First, the local hyperfine interaction at a P1 site (see Eq. 5) creates an extra field (\( +A_1, -A_1 \), or zero), which is determined by the nitrogen’s nuclear spin \( I_z^k \); the magnitude of this field also depends on the type of the P1 center, since \( A_1 \) is different for the centers of type 1 and the centers of type 2, 3, and 4. Thus, for the \( j \)-th bath spin to flip, it is necessary that the \( k \)-th spin has the value of \( I_z^k \) equal to \( I_z^j \), and that these two centers have the same \( A_1 \). Second, at large magnetic field, the internal Hamiltonian of the bath is renormalized, see Eq. 8, and different extra fields, created by the NV center, act on different P1 centers. As a result, the dynamics of the bath spins at large magnetic field is strongly suppressed, and only a few spins can change their orientation with time.

V. ANALYTICAL THEORY FOR DECOHERENCE DYNAMICS OF AN NV CENTER

Exact microscopic calculations, based directly on Eqs. 2, 7, and 9 for zero magnetic field (or Eqs. 3, 8, and 9) for large magnetic field) are too complex for analytics, and we use numerical simulations described in Sec. VI. For analytical treatment, a simplified model
is needed. Below, we provide justification, and describe such a model. We consider the relevant experiments (decay of Ramsey fringes, damping of Hahn spin echo, and decay of Rabi oscillations), and show how our analytical model helps in understanding the results.

A. Description of the phenomenological model: justification and qualitative conclusions

We start from summarizing the results of the microscopic considerations given above. For zero magnetic field, the Hamiltonian of the NV center coupled to the bath of nitrogen atoms, has a form

\[ H = B_z S_z^0 + h_x S_x^0 + H_B \] (11)

where \( H_B \) is the internal Hamiltonian of the bath (it includes both local Hamiltonians of the P1 centers and the dipolar interactions between them), \( h_x = (1/2)g_0\mu_B H_R \), and \( B_z = A_0 I_0^z + \tilde{a}_k \tilde{S}_k^z \). For large magnetic field, the relevant Hamiltonian has a similar form

\[ H = B_z s_0^z + h_x s_0^x + H_B \] (12)

where \( h_x = (1/\sqrt{2})g_0\mu_B H_R \), and \( B_z = A_0 I_0^z + c_k S_k^z \). Aside from the parameter values, the only difference between these two Hamiltonians is that the central spin \( S_0 = 1 \) at zero magnetic field while \( s_0 = 1/2 \) at large magnetic field.

If we omit for a moment the bath’s internal Hamiltonian \( H_B \), the exact solution of the problem can be obtained by purely analytical means for both zero and large magnetic field. It is important to recognize that \( I_0^z \) is a constant of motion, and, if \( H_B = 0 \), \( S_k^z \) (or \( \tilde{S}_k^z \)) are the constants of motion, too. Thus, if the \( k \)-th bath spin was initially in the state \( |\uparrow \rangle \), then it will remain in this state, and the operator \( S_k^z \) in this case can be replaced by a c-number equal to \( \langle \uparrow | S_k^z | \uparrow \rangle = 1/2 \). Thus, \( B_z \) in Eqs. 11 and 12 can be treated simply as some real number, whose value depends upon the specific initial state of the bath. Finally, we must average over all possible initial states of the bath; all these states are equally probable (at room temperature). The corresponding distribution \( P(B_z) \) of the values of \( B_z \) is given by the central limit theorem of the probability theory: it is a Gaussian distribution, with the mean equal to \( A_0 I_0^z \) (i.e., to \(+A_0\), \(-A_0\), or zero, with equal probability), and with the variance \( \tilde{b}^2 = (1/4) \sum_k c_k^2 \) (or, for zero magnetic field, \( \tilde{b}^2 = (1/4) \sum_k \tilde{a}_k^2 \)). Thus, for \( H_B = 0 \),
it is a rigorously justified step to replace the action of the static bath by the action of the static Gaussian random field $B_z$.

In reality, when the internal bath dynamics determined by $H_B$ is not negligible, the bath spins can change its orientation (flip), and $S^z_k$ is not an integral of motion anymore. In order to deal with this complication, we make an important approximation: we treat the bath in a mean-field manner, assuming that the influence of the bath spins on the NV center can still be approximated as the action of some random field $B_z$, which now may depend on time. This mean-field approximation is a standard (and probably, the most transparent) way for describing a wide variety of NMR/ESR-like experiments, successfully used since 1950s [3, 4] till present times [5]. Of course, applicability of the mean-field approximation requires an independent confirmation. In our case, the validity of the model is confirmed by direct microscopic numerical simulations, and by very good agreement with the experimental results. This is also the reason why, in this work, we do not use more sophisticated approaches, which, for instance, take into account two-spin correlations inside the bath [5]. Such approaches are less qualitatively clear than the mean field, but, unlike direct numerical simulations, still need to invoke assumptions about the bath dynamics (e.g., that the 2-spin correlations are sufficient); checking validity of these assumptions and development of such theories for NV centers requires a separate detailed investigation.

In order to construct the correct model for the dynamics of the random field $B_z$, we recall (see Sec. IV C) that the rate of flipping of a given nitrogen spin depends very strongly on the type of this nitrogen atom, on the value of the parameter $F$ (for zero magnetic field) or the value of $I^z_k$ of this atom (for large magnetic field), and on the state of its neighbors. Therefore, for some spins $S^z_k$ can be well approximated as static, while for other spins this approximation is invalid. Moreover, whether $S^z_k$ can be approximated as static depends on the coupling of the $k$-th spin to the NV spin. This fact, derived during early days of the NMR/ESR theory [6], can be qualitatively explained as follows (for formal derivation, see the original work, Ref.[6]). Let us assume that the characteristic time for a flip of a given spin is $\tau_c$, and the coupling constant is $\Delta_k$. This spin imposes a random time-varying field on the NV center, so that, after some time $t$, the state $|m_s = +1\rangle$ of the NV center acquires some phase $\beta$, while the state $|m_s = -1\rangle$ acquires a phase $-\beta$. The quantity determining the decoherence rate is the average of exp ($i\beta$) over all possible realizations of the random field; in Ref.[6], this average is denoted as $v_k(t)$. The value of $v_k$ depends on
whether \( \Delta_k \tau_c \gg 1 \) or \( \Delta_k \tau_c \ll 1 \): in the former case, the \( k \)-th bath spin is practically static, and \( v_k(t) \approx 1 - \Delta_k^2 t^2/2 \), while in the latter case, the \( k \)-th spin is in the regime of motional narrowing [3, 4] with \( v_k(t) \approx \exp(-\Delta_k^2 \tau_c t/2) \).

In a real bath, there are many spins with different \( \Delta_k \) and different \( \tau_c \). We simplify the situation in the spirit of the work [6], assuming that the spins with \( \Delta_k \tau_c > 1 \) can be treated as static, while the spins with \( \Delta_k \tau_c < 1 \) can be treated as being in the regime of complete motional narrowing. Thus, the mean field \( B_z \) acting on the NV center contains two components: one is a static random Gaussian field with the mean \( A_0 I_0^z \) and the variance \( b^2 \) (or \( \tilde{b}^2 \), for zero magnetic field), and the other is the fast Gaussian random field with the mean zero, variance \( b_1^2 \) (or \( \tilde{b}_1^2 \)) and correlation time \( \tau_c \) such that \( b_1 \tau_c \ll 1 \) (or, for zero magnetic field, \( \tilde{b}_1 \tilde{\tau}_c \ll 1 \)).

Based on our knowledge of microscopic Hamiltonians (see discussions in this Section above and Sec. IV C), we can make some qualitative conclusions about expected experimental results. At large magnetic field, in comparison with the zero-magnetic field case, the majority of bath spins are difficult to flip, and the spin correlation time \( \tau_c \) at large magnetic field should be larger than the correlation time \( \tilde{\tau}_c \) at zero magnetic field. Moreover, the coupling coefficients \( \tilde{a}_k \) at zero magnetic field are, in general, smaller than the coupling coefficients \( a_k \) at large magnetic field. Indeed, at zero magnetic field, due to the hyperfine coupling between the electron and nuclear spins of a given nitrogen atom, the matrix elements of the operators \( S_k^{x,y,z} \) are significantly smaller than 1/2, see Eqs. 7), so that the values of the coupling coefficients \( \tilde{a}_k \) are, in average, smaller. At large magnetic field, no such renormalization exist, and the coupling coefficients \( a_k \) are, in average, larger than \( \tilde{a}_k \). Thus, at large magnetic field, more spins satisfy the inequality \( \Delta_k \tau_c > 1 \), i.e., at large magnetic field the majority of spins is static. Therefore, the value of \( b \) should be larger than \( \tilde{b} \), while \( b_1 \) should be smaller than \( \tilde{b}_1 \). This is exactly what we observe in experiments. Furthermore, we expect that, qualitatively, the Ramsey fringes decay should be faster at large magnetic fields (where the static component of the bath field is larger), while the decay of Hahn spin echo should be faster at zero magnetic field (where the dynamic component of the bath field is larger). Again, this is what we observe in experiments.

Here, we do not go beyond these qualitative conclusions: development of a detailed microscopic theory for the bath parameters \( (b \) and \( \tilde{b} \), \( b_1 \) and \( \tilde{b}_1 \), etc.) is not a main topic of our work. Such a theory would involve many complications: the bath spins in our case are
complex entities, the interaction coefficients have complex dependence on the internal state of the nitrogen atoms and on the position of a given nitrogen atom, etc. (e.g., note that in the work [6] the dependence of $\Delta_k$ on the direction of the vector $n_k$ is neglected). A detailed theory of this kind has been developed, e.g., for decoherence of a spin-1/2 P donor by a bath of $^{29}$Si spins 1/2 [5], but even for that simpler bath the description is very complex. Development of similar description for NV centers would require a separate investigation. Therefore, everywhere below we treat the bath parameters as phenomenological quantities, which are extracted from experiments.

B. Analytical solution for the Ramsey fringes experiments

As a simplest example, let us consider the decay of Ramsey fringes; this experiment is equivalent to the free induction decay in NMR/ESR. The NV center is prepared in the state $|m_S = 0\rangle$, and a $\pi/2$ pulse is applied along the $x$-axis. We assume that the $\pi/2$ rotation is ideal (the width of the pulse is of order of 10 ns, and the influence of the field $B_z$ during this interval can be neglected). At zero magnetic field, the initial state is

$$|\psi_0\rangle = -\frac{i}{\sqrt{2}} (|m_S = 1\rangle + |m_S = -1\rangle).$$

(13)

In the absence of driving ($h_x = 0$), the evolution of this state is easy to calculate: after time $t$, the state $|m_S = 1\rangle$ acquires a phase $\beta = \int_0^t B_z(s) ds$, while the state state $|m_S = -1\rangle$ acquires a phase $-\beta$. Then, another $\pi/2$ pulse is applied, and the population $p_0(t)$ of the state $|m_S = 0\rangle$ is measured, which is equal to $p_0(t) = (1 + \cos 2\beta)/2$; this value should be averaged over all possible $\beta$ to give the experimentally detected average value $\langle p_0(t) \rangle$.

In our model, $B_z$ is a sum of two components, static $B_z^s$ and dynamic $B_z^d(t)$, and their contributions to $\beta$ are additive. Therefore, we can first perform averaging over the static component, obtaining

$$\langle p_0(t) \rangle^s = \frac{1}{2} + \frac{1}{6} [1 + 2 \cos 2A_0t] \exp (-2\tilde{b}^2 t^2),$$

(14)

where $\langle \cdot \rangle^s$ reminds that an averaging over $B_z^d(t)$ still should be performed. For large magnetic field, after the $\pi/2$ pulse, the state of the NV center is $(1/\sqrt{2})[|m_S = 0\rangle - i|m_S = -1\rangle]$. The calculations of $\langle p_0(t) \rangle^s$ can be done in a similar way, and the answer is:

$$\langle p_0(t) \rangle^s = \frac{1}{2} - \frac{1}{6} [1 + 2 \cos A_0t] \exp (-\tilde{b}^2 t^2/2).$$

(15)
Note that for zero magnetic field, \( \langle p_0(0) \rangle = 1 \), since two subsequent \( \pi/2 \) pulses bring the state \( |m_S = 0\rangle \) back to \( |m_S = 0\rangle \), while for large magnetic field, two \( \pi/2 \) pulses transfer \( |m_S = 0\rangle \) to \( |m_S = -1\rangle \). The free induction decay (FID) given by Eqs. 14 and 15 has Gaussian form \( \exp \left( -t^2 \right) \).

Now, we should perform an average over the dynamic component \( B_d^z(t) \). This calculation is a part of standard NMR/ESR theory [3, 4]. For our problem, it can be shown that the averaging over \( B_d^z(t) \) leads simply to multiplying the \( \langle p_0(t) \rangle^s \) by a factor \( \exp \left( -b_1^2 \tau_c t/2 \right) \) (or \( \exp \left( -\tilde{b}_1^2 \tilde{\tau} t/2 \right) \)). In the next subsection, we show that this is exactly the factor which determines the attenuation of the Hahn spin echo. Thus, expressing the decay of Ramsey fringes in terms of experimentally measured quantities, we have:

\[
\langle p_0(t) \rangle = \frac{1}{2} + \frac{1}{6} \left[ 1 + 2 \cos 2A_0 t \right] \exp \left[ -2\tilde{b}^2 t^2 \right] \exp \left( -t/T_2 \right),
\]

for zero magnetic field, where \( T_2 \) denotes the decay time of the Hahn spin echo at zero magnetic field, and

\[
\langle p_0(t) \rangle = \frac{1}{2} - \frac{1}{6} \left[ 1 + 2 \cos A_0 t \right] \exp \left( -\tilde{b}^2 t^2 /2 \right) \exp \left( -t/T_2 \right),
\]

for large magnetic fields, where \( T_2 \) is the decay time of the Hahn spin echo at large magnetic field.

### C. Analytical results for Hahn spin echo decay

In the Hahn spin echo experiments, after applying the first \( \pi/2 \) pulse, we let the system evolve during the time interval \( \tau \), then apply the \( \pi \) pulse, wait for another time \( \tau \), and then apply the second \( \pi/2 \) pulse followed by the readout of \( p_0(t) \). To analyze the Hahn spin echo theoretically, we note that during the intervals of free evolution between the pulses, the contributions to the phase \( \beta \) from \( B_s^z \) and \( B_d^z \) are additive, and can be analyzed separately. The effect of the static component is removed completely by the \( \pi \) pulse: the phase acquired due to \( B_s^z \) during first interval \( \tau \) is equal in magnitude, and opposite in sign, to the phase acquired due to \( B_s^z \) during the second interval \( \tau \).

Calculation of the effect of the dynamic component is also a standard part of NMR/ESR theory [3, 4], and the answer is rather expected: for the extreme motional narrowing regime, the decay of the Hahn spin echo is the same as the free induction decay (Ramsey fringes...
decay); denoting \( t = 2\tau \), we obtain:

\[
\langle p_0(t) \rangle = \frac{1}{2} + \exp \left( -\hat{b}_1^2 \tilde{\tau}_c t / 2 \right) = \frac{1}{2} + \exp \left( -t / \tilde{T}_2 \right),
\]

(18)

for the zero magnetic field, and

\[
\langle p_0(t) \rangle = \frac{1}{2} + \exp \left( -b_1^2 \tau_c t / 2 \right) = \frac{1}{2} + \exp \left( -t / T_2 \right),
\]

(19)

for large magnetic field. Indeed, in the motional narrowing regime, the bath fluctuations are fast, and the effect of the \( \pi \) pulse is quickly forgotten. Note that the measurements of the Ramsey fringes and Hahn spin echo decay do not allow reliable determination of both \( b_1 \) and \( \tau_c \) (or \( \tilde{b}_1 \) and \( \tilde{\tau}_c \)): only their combination \( b_1^2 \tau_c \) determines the observable quantity \( T_2 \).

\*\*\*\*\*\*\*\*\*

**D. Rabi oscillations**

Theoretical analysis of the Rabi oscillations is facilitated if performed in the rotating frame, which rotates with the frequency of the driving field \( H_R \); in this way, the NV center Hamiltonian is static, see Sec. IIIA and IIIB. Furthermore, in order to observe well-defined Rabi oscillations, a rather large driving field \( H_R \) is needed: otherwise, the decay time of the oscillations will be of the same order as an oscillation period. Thus, we perform analysis assuming that \( h_x \gg B_z \) and \( h_x \tau_c \gg 1 \). These assumptions are not unavoidable: it is possible also to study analytically other situations with slowly decaying oscillations, but the results of different calculations agree with each other, while the approximation of large \( h_x \) allows to give a clear qualitative picture of underlying physics.

First, we note that, for the purposes of the present paper, the influence of the dynamical component of \( B_z \) can be neglected. Qualitatively, it is a very natural result: the Rabi rotation of the spin \( S_0 \) around \( h_x \) is the fastest process, which averages out the field \( B_z \). If the dynamic part of \( B_z \) is sufficiently slow, much slower than the rotation of \( S_0 \) (i.e., \( h_x \tau_c \gg 1 \)), then it just introduces an extra averaging, which is small in comparison with the averaging due to fast Rabi rotation. This conjecture can be confirmed by explicit calculations within the random-field approximation [12], but these calculations are very lengthy and complex to reproduce here, and are far beyond the scope and the main focus of the present paper. For the purposes of the present paper, we just note that neglecting the dynamic part of \( B_z \) is confirmed as a good approximation by the agreement with the numerical simulations (which do not neglect the bath dynamics) and with the experimental results.
At zero magnetic field, the relevant Hamiltonian is

$$H = B_z S_0^z + h_x S_0^x$$

(20)

with $B_z$ being a static Gaussian field with mean $A_0 I_0^z$ and variance $\tilde{b}$. The time evolution operator for this Hamiltonian can be found exactly for any given $B_z$:

$$U = \exp(-iHt) = 1 + (\cos \Omega t - 1) \left[ (2/3) + 2u_x u_z Q_x^x + u_z^2 Q_x^x + u_x^2 Q_z^z \right] - i [u_x S_0^x + u_z S_0^z] \sin \Omega t,$$

(21)

where $\Omega = \sqrt{B_z^2 + h_x^2}$, $u_x = h_x / \Omega$, $u_z = B_z / \Omega$, and $Q_{0}^{\alpha \beta} = (1/2)[S_0^\alpha S_0^\beta + S_0^\beta S_0^\alpha] - (2/3) \delta_{\alpha \beta}$ are the quadrupolar operators.

In the limit of $h_x \gg \tilde{b}$, for times larger than $\tilde{b}^{-1}$, one can show that it is sufficient to expand $\Omega$ up to second order in $B_z/h_x$, and neglect $B_z$ everywhere else (i.e., take $u_z = 0$ and $u_x = 1$). The probability $p_0(t)$ then has a very simple form:

$$p_0(t) = \cos^2 \Omega t$$

(22)

and should be averaged over all possible $B_z$, which has a distribution

$$P(B_z) = (1/3)(P_+ + P_- + P_0)$$

where $P_{+, -, 0}$ are all Gaussians with the variance $\tilde{b}$, and mean $+A_0$ for $P_+$, $-A_0$ for $P_-$, and zero for $P_0$. These three Gaussians correspond to $m_I = +1$, $m_I = -1$, and $m_I = 0$ respectively, and the average $\langle p_0(t) \rangle$ is an arithmetic average of three contributions. The contribution from $m_I = 0$ part is a slowly decaying phase-shifted oscillation:

$$\langle p_0(t) \rangle_0 = \frac{1}{2} + \frac{1}{2} \left[ 1 + 4 \frac{h_x^2 t^2}{h_x^2} \right]^{-1/4} \cos (2h_x t + \phi)$$

(23)

where the phase shift is $\phi = (1/2)\arctan(2b^2 t/h_x)$. Note that the decay at long times is of power-law form $\propto 1/\sqrt{t}$, and has noticeable amplitude even at very large times. Such an interesting decay has been noticed in earlier works [13] (see also References 8-10 of the main text), and signatures of this behavior have been recently observed in quantum dots [14].

The contributions from the parts $m_I = +1$ and $m_I = -1$ are equal to each other, and have a slightly more complex form:

$$\langle p_0(t) \rangle_{\pm} = \frac{1}{2} + (1+\xi^2)^{-1/4} \exp \left[ -(1/2)v^2 \xi^2 / (1 + \xi^2) \right] \cos [2h_x t + v^2 \xi + \phi - (1/2)v^2 \xi^3 / (1 + \xi^2)]$$

(24)

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where we introduced the "dimensionless time" $\xi = 2\tilde{b}^2t/h_x$, the quantity $v = A_0/b$, and the phase $\phi = (1/2)\arctan \xi = (1/2)\arctan(2\tilde{b}^2t/h_x)$ is the same as above.

Inspection of Eq. 24 shows that $\langle p_0(t) \rangle_{\pm}$ exhibits a product of two decaying functions and one oscillating cosine with time-varying phase. One decaying function is the same power-law $1/\sqrt{t}$ decay (the term $(1 + \xi^2)^{-1/4}$ which appeared in Eq. 23, and the other one is the exponent which saturates at $t \to \infty$ at the value $\exp (-v^2/2)$. At zero magnetic field, when $\tilde{b} \ll A_0$, the Eq. 24 is considerably simplified: in this case $v \gg 1$, and all terms which do not involve $v$ (except for the large phase $2h_xt$) can be neglected. The result is a simple Gaussian decay of oscillations

$$\langle p_0(t) \rangle_{\pm} = \frac{1}{2} + \frac{1}{2} \exp \left[ -2A_0^2\tilde{b}^2t^2/h_x^2 \right] \cos 2h_xt$$

with the characteristic decay time $h_x/(A_0\tilde{b})$.

This result is very natural (see also Figure 3 and accompanying part of the main text). For $m_I = 0$, the Rabi field $h_x$ is perpendicular to the $B_z$ field, and the fast rotation of $S_0$ around $h_x$ completely averages out $B_z$ in the first order, so that the effect of $B_z$ is seen only in the second order (the decay time in Eq. 23 is of order of $\tilde{b}^2/h_x$), and this effect is small, leading to a slow power-law decay. For $m_I = \pm 1$, due to the finite mean $\pm A_0$, the Rabi field $h_x$ is not exactly perpendicular to $B_z$, and the part of $B_z$ which is not averaged by fast Rabi rotation leads to a standard Gaussian decay.

When we sum the contributions from all three values of $m_I$, an interesting effect takes place. The oscillations of $\langle p_0(t) \rangle_0$ decay very slowly, but have a time-varying phase, while the oscillations of $\langle p_0(t) \rangle_{\pm}$ decay rather fast. Thus, at some moment of time, the term $\langle p_0(t) \rangle_0$ can be equal to twice $\langle p_0(t) \rangle_{\pm}$ but have an opposite sign. In this case, the total $\langle p_0(t) \rangle$ would be equal to zero. Correspondingly, Rabi oscillations around that time would demonstrate a dip. Later, as $\langle p_0(t) \rangle_{\pm}$ decay further, this dip would disappear, reviving the Rabi oscillations.

The model has two free parameters for the Rabi oscillations (dynamical spin bath components are neglected), the driving field and the width of the dipolar field distribution $b$. In order to compare the model to a given Rabi oscillation experiment, we determine the driving field from the frequency of the Rabi oscillations at large pulse widths. The width $b$ is determined independently from a Ramsey experiment. As shown in Figure 3 of the main text, the main features of the experiment are reproduced in detail by the model. In Figures
S2 and S3 the dependence on the driving frequency is investigated. The experimentally observed shift of the collapse to longer pulse widths for higher driving frequencies is also seen in the analytical model. Small deviations remain between model and experiment, such as the slightly faster long-time decay in the experiment. This may be explained by the dynamical components of the bath, which are neglected in the model. The fact that the numerical simulations, which include the dynamical components, are closer to the experiment in this respect seems to confirm this.

At large magnetic field, when the NV center is described by a pseudo-spin $s_0 = 1/2$, the analysis can be performed in exactly the same way. In this case, one can show that for a given $B_z$,

$$p_0(t) = \cos^2 \Omega t/2$$

and, up to replacing $t \rightarrow t/2$, is the same as in the zero-magnetic field case, Eq. 22. Thus all results above remain the same, with obvious modification $\tilde{b} \rightarrow b$. In particular, the contribution of $m_I = 0$ has the same form of slowly decaying $(1/\sqrt{t})$ oscillations with the time-varying phase. However, at large magnetic fields $A_0 \sim b$, and the contributions of $m_I = \pm 1$ do not differ crucially from the contributions of $m_I = 0$. Instead, the parts of the $P(B_z)$ distribution which are closer to zero have smaller value of $\nu$, and their decay is more of a power-law form, while the parts which are farther from zero have larger value of $\nu$, and their decay is faster, closer to the fast Gaussian decay. The corresponding experiment is shown in Figure 4.

A few remarks are in order:

First, we note here that the Eqs. 23 and 25 can be obtained (with insufficient modifications) also in other cases, e.g. when $h_x \sim A_0$, $h_x \gg b$. The derivation above was chosen because only because it is physically more transparent, and allows to discuss the zero-magnetic field and the large-magnetic field cases on equal footings.

Second, it is worthwhile to note that exactly the same results can be obtained if, instead of the random field $B_z$, we had considered the full quantum bath (i.e., considered $B_z$ as an operator) but assuming that the bath is static ($H_B = 0$). Consequently, in case of Rabi oscillations, again, the full quantum static bath is rigorously equivalent to static random field.

Finally, we briefly comment on the Rabi oscillations at 514 G, which demonstrate interesting two-frequency pattern. At this magnetic field, the frequency of the Rabi driving field
equals the transition frequency of both NV center, and the nitrogen atoms. However, the term describing the response of the NV center to the Rabi field (in the rotating frame) is of the form \((1/\sqrt{2})g_0 \mu_B H_R s_0^x\), see Eq. 3. At the same time, the response of the nitrogen spins to the Rabi field is \(\sqrt{2}\) times smaller. This corresponds to the experimental observations: the second Rabi frequency of the NV center is \(\sqrt{2}\) times smaller than its ”native” frequency, and corresponds to the Rabi frequency of the nitrogen atoms. The fact that the second frequency is present at very early times, of order of 0.1 \(\mu\)s, strongly suggests that already at zero time, an entanglement between the NV center and the nitrogen spins is present. The detailed description of this entangled state and its dynamics is currently under development.

VI. MICROSCOPIC THEORY: NUMERICAL SIMULATIONS

The numerical simulations have been performed in the rotating frame, starting directly from the Hamiltonians given by Eqs. 2, 7, and 9 for zero magnetic field, and Eqs. 3, 8, and 9 for large magnetic field. Thus, the nuclear spins of the NV center and of the nitrogen P1 centers have been explicitly taken into account, as well as the dipolar interactions between the nitrogen spins.

The NV center was placed at the origin of the coordinate frame, and the nitrogen atoms have been randomly placed around the NV center. The distances to the nitrogen atoms have been all scaled by the same factor, to give the experimentally measured decay time of Ramsey fringes; this scaling factor is the only adjustable parameter in the numerical simulations. In this way, we obtained the correct local density of bath spins. Note that we did not average over positions of the nitrogens: such an averaging would correspond to the ensemble of the NV centers, and, as we discussed in Sec. V B, would not be correct for a measurement of a single NV center.

The state of the system was described by an exact many-spin wavefunction, including both the NV center and the nitrogen atoms. Initially, the state of the NV center was \(|m_S = 0\rangle\), while the bath spins were in a random superposition of all possible bath basis states. This superposition state gives an excellent (exponentially accurate) approximation for the experimentally relevant state of the bath (maximally mixed, with the density matrix proportional to the identity matrix) [15].

The dynamics of the system was simulated using an expansion of the time-evolution
operator $\exp(-itH)$ in terms of Chebyshev polynomials [15]. The number of nitrogen atoms was varied from $N = 4$ to $N = 6$. Simulations of larger systems is rather demanding, since the total number of basis states is rather large. For a single nitrogen atom, we need six basis states (two electron spin states, three nuclear spin states), so that the simulations with $N = 6$ involve 140,000 basis states ($\simeq 47,000$ basis states for the bath, multiplied by three basis states for the NV spin). At the same time, the simulation timestep should be kept small due to large value of the Rabi driving field $H_R$, so that the total number of timesteps is rather large.

Moreover, we took into account the fact that each nitrogen can have different directions of the delocalization axis. The directions of the delocalization axes were randomly assigned for all nitrogens, and the averaging over 10 realizations was taken. Also, we performed averaging over three possible $z$-projections of the nuclear spin of the NV center; this projection is a constant of motion.

The simulations have been performed for all three types of relevant experiments: Ramsey fringes decay, Hahn spin echo decay, and damping of the Rabi oscillations. Figure S4 shows simulations of the Hahn spin echo and the Ramsey experiment, for the same configuration of the P1 centers as in the simulation shown in Figure 3. Fitting the simulations with our model equations yields values for both $T_2$ and $b$ that agree with those of NV14 within error bars. Therefore, this configuration is a good representation of the environment of NV14, which is also reflected in the agreement between experiment and simulation in the Rabi oscillations.

[12] V. V. Dobrovitski et al. (to be published).
Supplementary Figure Captions

Figure S1. Spin echo measurements.
(A) Pulse sequence for the spin echo measurements. (B) Spin echo signal of NV31 at $B = 0$ G as a function of $\tau_2$ for different $\tau_1$. (C) Measurement of coherence time $T_2$ of NV31 using the spin echo pulse scheme of (A) with $\tau_1 = \tau_2 = \tau$. Fitting the data to $\exp(-2\tau/T_2)$ yields $T_2 = (0.62 \pm 0.09) \mu$s at $B = 0$ G and $T_2 = (5.0 \pm 0.5) \mu$s at $B = 740$ G.

Figure S2. Comparison of experiment with analytical model for Rabi oscillations of NV14 at $B = 0$ G.
Experimental Rabi oscillations of NV14 (top panels) and calculations using our analytical model (bottom panels) with $b = 0.42$ MHz, for driving frequencies of (A) 11.7 MHz, (B) 21.4 MHz, and (C) 25.6 MHz.

Figure S3. Comparison of experiment with analytical model for Rabi oscillations of NV31 at $B = 0$ G.
Experimental Rabi oscillations of NV31 (top panels) and calculations using our analytical model (bottom panels) with $b = 0.47$ MHz, for driving frequencies of (A) 12.1 MHz, (B) 15.3 MHz, and (C) 20.5 MHz.

Figure S4. Numerical simulation of spin echo and free evolution at $B = 0$ G.
Numerical simulations of the dynamics of a single NV spin surrounded by a bath of six nitrogen impurities at $B = 0$ G (see text). (A) Spin echo experiment. Fitting the numerical results yields $T_2 = (0.41 \pm 0.2) \mu$s. (B) Free evolution. Fitting the numerical results with the value for $T_2$ found from (A) gives $b = (0.37 \pm 0.02)$ MHz.
Figure S1. Hanson et al.
Figure S2. Hanson et al.
Figure S3. Hanson et al.
Figure S4. Hanson et al.