

Supplementary Information

Contents

1	Hamiltonian of the NV center	1
2	Detailed theory of the gradiometry protocol	2
2.1	Standard Gradiometry Sequence	2
2.2	Generalized sequence	4
2.3	Introducing a nuclear memory	6
2.4	Modified sequence for T_1 -limited gradiometry	8
3	Detailed description of the oscillating NV	9
4	Modelling of magnetic anomalies	10
5	Modeling of magnetic fields from neuron action potentials	11

1 Hamiltonian of the NV center

The nitrogen-vacancy (NV) center is a spin defect in diamond which comprises a substitutional nitrogen and a vacancy defect. In its negative charge state, this defect is an electron spin-1 system described by the following Hamiltonian:

$$\mathcal{H}_{NV} = \underbrace{DS_z^2 - \gamma_e B_0 S_z}_{\mathcal{H}_0} + \underbrace{A_{\parallel} S_z I_z + A_{\perp} (S_x I_x + S_y I_y)}_{\mathcal{H}_{hf}} + \underbrace{Q I_z^2}_{\mathcal{H}_Q} - \underbrace{\gamma_n B_0 I_z}_{\mathcal{H}_Z^{(n)}}. \quad (1)$$

Here, $D/(2\pi) = 2.88$ GHz is the zero-field splitting, B_0 is an external magnetic field aligned the NV quantization axis and $\gamma_e = -28.024$ GHz/T is the electron gyromagnetic ratio. The term $\mathcal{H}_{hf}$ describes the hyperfine interaction between the electron spin-1 $\mathbf{S}$ and the nitrogen nuclear spin $\mathbf{I}$, where $A_{\parallel}/(2\pi) = -2.2$ MHz for ^{14}N and $A_{\parallel}/(2\pi) = -3.1$ MHz for ^{15}N , and $A_{\perp}$ can be neglected due to the large zero-field splitting of the center. For a ^{15}N nucleus with spin $I = 1/2$, the quadrupolar interaction $\mathcal{H}_Q$ disappears, while $Q/(2\pi) = -4.945$ MHz for a ^{14}N nucleus with spin $I = 1$. The last term in Eq. (1) is the nuclear Zeeman interaction due to the external field B_0 , where $\gamma_{^{14}\text{N}}/(2\pi) = 3.077$ MHz/T and $\gamma_{^{15}\text{N}}/(2\pi) = -4.316$ MHz/T. Throughout the text, we consider a ^{14}N nuclear host for the NV center.

A microwave (MW) driving field can be applied to control the electron spin (e) of the NV center,

$$\mathcal{H}_{\text{MW}}(t) = \sqrt{2}\Omega_{\text{MW}} \cos(\omega_{\text{MW}}t + \phi_{\text{MW}}) S_x, \quad (2)$$

where ω_{MW} (Ω_{MW}) is the (Rabi) frequency of the driving and ϕ_{MW} its initial phase. Tuning the driving frequency to match the lower energy transition $|0\rangle_e \rightarrow |\bar{1}\rangle_e$ induces population transfer between these levels, such that the NV center can be effectively treated as a spin-1/2 system. Note that, in this scenario, $S_z = (\sigma_z - \mathbb{1})/2$, with σ_z the third Pauli matrix (in the basis $\{|0\rangle, |\bar{1}\rangle\}$). Moving to the rotating frame with respect to $\mathcal{H}_0$ and invoking the rotating

wave approximation, the MW control Hamiltonian (2) transforms into

$$\mathcal{H}_{\text{MW}}^I = \Omega_{\text{MW}} S_{\phi_{\text{MW}}}, \quad (3)$$

where $S_{\phi_{\text{MW}}} = \sigma_{\phi_{\text{MW}}}/2$ and $\sigma_{\phi_{\text{MW}}} = \cos \phi_{\text{MW}} \sigma_x + \sin \phi_{\text{MW}} \sigma_y$, with σ_x and σ_y the first and second Pauli matrices. The propagator associated to the pulse Hamiltonian (3), $U_{\phi_{\text{MW}}} = e^{-i\mathcal{H}_{\text{MW}}^I t} = e^{-i\frac{\Omega_{\text{MW}} t}{2} \sigma_{\phi_{\text{MW}}}}$, describes a rotation of an angle $\theta_{\text{MW}} = \Omega_{\text{MW}} t/2$ around the axis set by ϕ_{MW} of the NV state in the Bloch state.

Similarly, a radio-frequency (RF) driving field may be used to control the nuclear spin (a) of the center:

$$\mathcal{H}_{\text{RF}}(t) = \Omega_{\text{RF}} \cos(\omega_{\text{RF}} t + \phi_{\text{RF}}) I_x, \quad (4)$$

where ω_{RF} (Ω_{RF}) is the (Rabi) frequency of the driving and ϕ_{RF} its initial phase. Tuning ω_{RF} to match the transition $|0\rangle_a \rightarrow |1\rangle_a$, Hamiltonian (4) becomes

$$\mathcal{H}_{\text{RF}} = \Omega_{\text{RF}} I_{\phi_{\text{RF}}}, \quad (5)$$

with $I_{\phi_{\text{RF}}} = \sigma_{\phi_{\text{RF}}}/2$.

Then, in the following we will work in the two qubit basis, $\{|0\rangle_e|0\rangle_a, |0\rangle_e|1\rangle_a, |\bar{1}\rangle_e|0\rangle_a, |\bar{1}\rangle_e|1\rangle_a\}$, and initialize the system in the $|0\rangle_e|0\rangle_a$ state:

$$\rho_0 = |0\rangle\langle 0|_e \otimes |0\rangle\langle 0|_a = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}. \quad (6)$$

2 Detailed theory of the gradiometry protocol

2.1 Standard Gradiometry Sequence

Our gradiometry sequence comprises three main stages: two interrogation stages and a signal evolution stage (see Fig. 1). During the first interrogation stage (I), a $(\pi/2)_y$ -pulse takes the sensor state to a superposition. Then, the sensor accumulates some phase ϕ_0 due to a quasi-static magnetic field $B(t)$. Next, a $(\pi/2)_x$ -pulse encodes the coherence information into qubit populations, such that the state of the sensor reads:

$$\rho_0 = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \rightarrow \rho_I = \frac{1}{2} \begin{pmatrix} 1 + \sin \phi_0 & \cos \phi_0 \\ \cos \phi_0 & 1 - \sin \phi_0 \end{pmatrix}. \quad (7)$$

A stage of signal evolution (II) follows: the sensor accumulates a phase $\tilde{\phi}_1$ due to the time varying field $B(t)$, and its state becomes:

$$\rho_{II} = \frac{1}{2} \begin{pmatrix} 1 + \sin \phi_0 & e^{-i\tilde{\phi}_1} \cos \phi_0 \\ e^{i\tilde{\phi}_1} \cos \phi_0 & 1 - \sin \phi_0 \end{pmatrix}. \quad (8)$$

During the second interrogation stage (III), the field becomes quasi-static again, although its value might different than that of stage I due to the intermediate signal evolution stage. The

sensor accumulates a phase ϕ_1 during this interrogation stage and, at the end of the sequence, its state is written as

$$\rho_{III} = \frac{1}{2} \begin{pmatrix} 1 + \sin \phi_1 \cos \tilde{\phi}_1 \cos \phi_0 - \cos \phi_1 \sin \phi_0 & i \sin \tilde{\phi}_1 \cos \phi_0 + \cos \phi_1 \cos \tilde{\phi}_1 \cos \phi_0 + \sin \phi_1 \sin \phi_0 \\ -i \sin \tilde{\phi}_1 \cos \phi_0 + \cos \phi_1 \cos \tilde{\phi}_1 \cos \phi_0 + \sin \phi_1 \sin \phi_0 & 1 - \sin \phi_1 \cos \tilde{\phi}_1 \cos \phi_0 + \cos \phi_1 \sin \phi_0 \end{pmatrix}. \quad (9)$$

Upon readout of the sensor state, we find

$$\langle \sigma_z \rangle = \text{Tr}(\rho_{III} \sigma_z) = \sin \phi_1 \cos \tilde{\phi}_1 \cos \phi_0 - \cos \phi_1 \sin \phi_0, \quad (10)$$

which for small $\tilde{\phi}_1$ can be approximated as

$$\langle \sigma_z \rangle \approx \sin(\phi_1 - \phi_0). \quad (11)$$

For DC magnetometry, the target magnetic field $B(t)$ will typically comprise two parts: a static background field B and a slowly varying signal $b(t)$, which we want to detect, *i.e.*, $B(t) = B + b(t)$. Standard DC sensing techniques, such as the Ramsey sequence (stage I in Fig. 1) or, in the case of color centers, optically detected magnetic resonance (ODMR), rely on total intensity measurements, such that magnetic shielding from the environment is often required to suppress static fields or magnetic drifts. Our protocol intrinsically suppresses the effect of these, as long as the phase $\tilde{\phi}_1$ is small. Then, for a given measurement starting at time t_i , $\phi_1 - \phi_0 \propto (B + b_{III}^i) - (B + b_I^i) = b_{III}^i - b_I^i$, where $b_I^i = b(t_i)$ and $b_{III}^i = b(t_i + \Delta t)$, with $\Delta t = t_{II}$. Then, considering N consecutive differential measurements, the discretized temporal

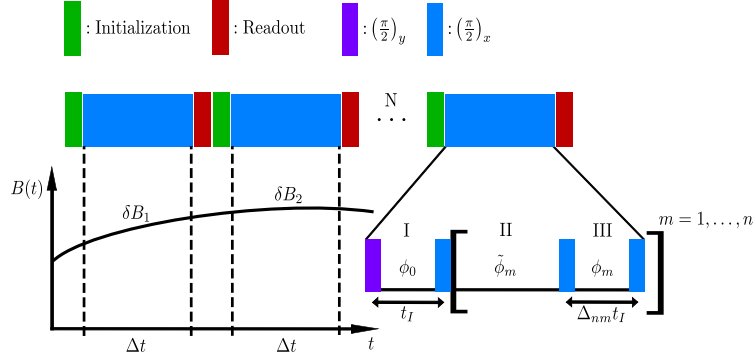


Fig. 1 Gradiometric sensing scheme. Our protocol comprises an initialization and a readout step, and a measurement sequence (in green, red and blue, respectively), sequentially repeated N times. Our sequence allows to measure the magnetic field difference between two points separated by a time Δt . Considering that the magnetic field $B(t)$ changes slowly within a single sequence, each measurement will be proportional to the field difference within the sequence, $\delta B_i = B(t_i + \Delta t) - B(t_i)$, such that, after N repetitions, the full gradient of the field can be recovered. Here, t_i sets the start of i -th measurement. Each of these sequences consists of two interrogation stages (I and III) wherein the value of the field remains unchanged, and a signal evolution stage (II) wherein the field changes its value. Repetition of stages II and III within each sequence allows access to n -th order derivative of the field. Here, the duration of stage III in the m -th block (relative to stage I) is given by the combinatorial number $\Delta_{nm} = \binom{n}{m}$.

gradient of the field can be reconstructed,

$$\left. \frac{dB(t)}{dt} \right|_{t_i} = \left. \frac{db(t)}{dt} \right|_{t_i} \approx \frac{\delta b_i}{\Delta t}, \quad (12)$$

where $\delta b_i = b_{III}^i - b_I^i$ is the magnetic field difference obtained in the i -th measurement stage (see Fig. 1).

2.2 Generalized sequence

Our gradiometric sequence can be generalized to measure higher order derivatives. By repeating stages II and III a total of n times, the n -th order derivative can be measured (see Fig. 1). We may write the sensor state at the end of the $(n-1)$ -th block as

$$\rho_{n-1} = \frac{\mathbb{1}}{2} + \frac{1}{2} \mathbf{v}_{n-1} \cdot \boldsymbol{\sigma}, \quad (13)$$

where $\mathbf{v}_{n-1} = (a_{n-1}, b_{n-1}, c_{n-1})$ is the Bloch vector and $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$. It is useful to consider how the Pauli matrices transform under rotations around different axes. For a $(\pi/2)$ rotation around the x axis, $\sigma_x \rightarrow \sigma_x$, $\sigma_y \rightarrow \sigma_z$ and $\sigma_z \rightarrow -\sigma_y$; and around the y axis, $\sigma_x \rightarrow -\sigma_z$, $\sigma_y \rightarrow \sigma_y$ and $\sigma_z \rightarrow \sigma_x$. For a φ rotation around the z axis, $\sigma_x \rightarrow \cos \varphi \sigma_x + \sin \varphi \sigma_y$, $\sigma_y \rightarrow \cos \varphi \sigma_y - \sin \varphi \sigma_x$ and $\sigma_z \rightarrow \sigma_z$.

Then, during stage II in the n -th block, the sensor accumulates a phase $\tilde{\phi}_n$ such that its state reads

$$\rho_{n-1}^{(II)} = \frac{\mathbb{1}}{2} + \frac{1}{2} \left[a_{n-1} (\cos \tilde{\phi}_n \sigma_x + \sin \tilde{\phi}_n \sigma_y) + b_{n-1} (\cos \tilde{\phi}_n \sigma_y - \sin \tilde{\phi}_n \sigma_x) + c_{n-1} \sigma_z \right]. \quad (14)$$

Next, the sensor undergoes the n -th stage III, during which it accumulates a phase ϕ_n , and the state before measurement reads

$$\begin{aligned} \rho_{n-1}^{(III)} = \frac{\mathbb{1}}{2} + \frac{1}{2} \left[(\cos \tilde{\phi}_n \cos \phi_n a_{n-1} - \sin \tilde{\phi}_n \cos \phi_n b_{n-1} + \sin \phi_n c_{n-1}) \sigma_x \right. \\ \left. + (-\sin \tilde{\phi}_n a_{n-1} - \cos \tilde{\phi}_n b_{n-1}) \sigma_y \right. \\ \left. + (\cos \tilde{\phi}_n \sin \phi_n a_{n-1} - \sin \tilde{\phi}_n \sin \phi_n b_{n-1} - \cos \phi_n c_{n-1}) \sigma_z \right] = \rho_n, \end{aligned} \quad (15)$$

from where we extract recursive relations for the components of the Bloch vector:

$$\begin{aligned} a_n &= \cos \tilde{\phi}_n \cos \phi_n a_{n-1} - \sin \tilde{\phi}_n \cos \phi_n b_{n-1} + \sin \phi_n c_{n-1}, \\ b_n &= -\sin \tilde{\phi}_n a_{n-1} - \cos \tilde{\phi}_n b_{n-1}, \\ c_n &= \cos \tilde{\phi}_n \sin \phi_n a_{n-1} - \sin \tilde{\phi}_n \sin \phi_n b_{n-1} - \cos \phi_n c_{n-1}, \end{aligned} \quad (16)$$

with $a_0 = \sin \phi_0$, $b_0 = 0$, $c_0 = \cos \phi_0$. Trivially, we find that $\langle \sigma_z \rangle = \text{Tr}(\rho_n \sigma_z) = c_n$, which for small $\tilde{\phi}_n$ can be written as

$$\langle \sigma_z \rangle = c_n \approx (-1)^n \sin \left[\sum_{m=0}^n (-1)^m \phi_m \right]. \quad (17)$$

To obtain expression (17), we first consider the coefficients a_n , b_n and c_n under the approximation $\tilde{\phi}_n \approx 0$:

$$\begin{aligned} a_n &\approx \cos \phi_n a_{n-1} + \sin \phi_n c_{n-1}, \\ b_n &\approx 0, \\ c_n &\approx \sin \phi_n a_{n-1} - \cos \phi_n c_{n-1}. \end{aligned} \quad (18)$$

Then, c_n can be expanded as

$$\begin{aligned} c_n &= \sin \phi_n a_{n-1} - \cos \phi_n c_{n-1} \\ &= \sin(\phi_n - \phi_{n-1}) a_{n-2} + \cos(\phi_n - \phi_{n-1}) c_{n-2} \\ &= \sin(\phi_n - \phi_{n-1} + \phi_{n-2}) a_{n-3} - \cos(\phi_n - \phi_{n-1} + \phi_{n-2}) c_{n-3} \\ &= \dots \end{aligned} \quad (19)$$

where angle sum and difference trigonometric identities have been used. Easily, one realizes that, for odd n , we find

$$\begin{aligned} c_n &= \sin[\phi_n - \phi_{n-1} + \dots + \phi_1] a_0 - \cos[\phi_n - \phi_{n-1} + \dots + \phi_1] c_0 \\ &= \sin[\phi_n - \phi_{n-1} + \dots + \phi_1 - \phi_0] \\ &= (-1) \sin \left[\sum_{m=0}^n (-1)^m \phi_m \right], \end{aligned} \quad (20)$$

while for even n

$$\begin{aligned} c_n &= \sin[\phi_n - \phi_{n-1} + \dots - \phi_1] a_0 + \cos[\phi_n - \phi_{n-1} + \dots - \phi_1] c_0 \\ &= \sin[\phi_n - \phi_{n-1} + \dots - \phi_1 + \phi_0] \\ &= (+1) \sin \left[\sum_{m=0}^n (-1)^m \phi_m \right], \end{aligned} \quad (21)$$

which can be combined into Eq. (17).

Now, note that the duration of the m -th stage III in the n -th order protocol is chosen to be $t_{III,m}^{(n)} = \Delta_{nm} t_I$, where

$$\Delta_{nm} = \binom{n}{m} \quad (22)$$

is the combinatorial number. Then, Eq. (17) can be approximated as

$$\langle \sigma_z \rangle^{(n)} \approx \sum_{m=0}^n (-1)^{n+m} \phi_m \propto (-1)^n b_I + \sum_{m=1}^n (-1)^{n+m} \Delta_{nm} b_{III,m} = \delta b, \quad (23)$$

where b_I and $b_{III,m}$ are the values of the signal $b(t)$ during stage I and the m -th stage III, respectively. Again, we see that the background field B is suppressed. As before, by performing N of such measurements, the discretized n -th order time derivative of the total field

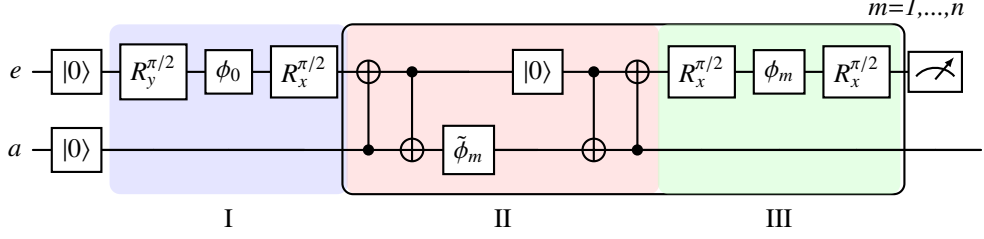


Fig. 2 General sequence for measuring the n -th order derivative of the field including an ancillary qubit serving as a nuclear memory. SWAP gates are used during stage II for encoding/recovering the sensor (e) state in/from the memory (a) such that the protocol is limited by the nuclear dephasing time T_2^a . Stages II and III are repeated n times, with the duration of the m -th repetition of stage III given by $t_{III,m}^{(n)} = \Delta_{nm} t_I$, where $\Delta_{nm} = \binom{n}{m}$.

$B(t) = B + b(t)$ can be obtained:

$$\left. \frac{d^{(n)} B(t)}{dt^n} \right|_{t_i} \approx \frac{\delta b_i}{\Delta t^n}, \quad (24)$$

with Δt the duration of stage II and the superscript i referring to the i -th measurement. As an example, consider the second order protocol; here, $\Delta_{21} = 2$ and $\Delta_{22} = 1$, so that we find

$$\langle \sigma_z \rangle^{(2)} \propto b_I - 2b_{III,1} + b_{III,2} = \delta b, \quad (25)$$

and thus

$$\left. \frac{d^{(2)} B(t)}{dt^2} \right|_{t_i} = \left. \frac{d^{(2)} b(t)}{dt^2} \right|_{t_i} \approx \frac{b_I^i - 2b_{III,1}^i + b_{III,2}^i}{t_{II}^2}, \quad (26)$$

which is the discretized second order derivative of the field $B(t)$.

2.3 Introducing a nuclear memory

The gradiometry protocol proposed works provided the phases $\tilde{\phi}_n$ are small, as the approximation in Eqs. (11) and (17) does not hold when the duration of the n -th signal evolution stage exceeds the sensor dephasing time T_2^* . Luckily, because we want this phase to have no effect on the final result, we can apply refocusing pulses such that this limitation is extended to the coherence time T_2 . However, this may not suffice if the target signal changes in a timescale larger than T_2 . In these cases, we may transfer the phase accumulation process of stage II to a nuclear ancillary qubit or nuclear memory through SWAP gates (see Fig. 2). These can easily be constructed by the consecutive application of CNOT gates.

A weak MW pulse ($\Omega_{MW} \ll A_{||}$) resonant with the $|0\rangle_e |1\rangle_a \rightarrow |\bar{1}\rangle_e |1\rangle_a$ transition results in a controlled-rotation operation, with the control being the memory and the target the sensor, *i.e.*, a $C_a \text{ROT}_e$ gate, which is described by

$$U_{C_a \text{ROT}_e}^{\phi_{MW}}(\theta_{MW}) = \mathbb{1} \otimes |0\rangle\langle 0|_a + e^{-i \frac{\theta_{MW}}{2} \sigma_{\phi_{MW}}} \otimes |1\rangle\langle 1|_a, \quad (27)$$

where the angle of rotation is $\theta_{MW} = \Omega_{MW} t_p$, t_p being the duration of the pulse, and ϕ_{MW} is the pulse axis. For the particular case where $\theta_{MW} = \pi$, the propagator (27) describes a

$C_a\text{NOT}_e$ -gate:

$$U_{C_a\text{NOT}_e} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & -i \\ 0 & 0 & 1 & 0 \\ 0 & -i & 0 & 0 \end{pmatrix}, \quad (28)$$

where we have taken $\phi_{\text{MW}} = 0$ for simplicity.

Similarly, a weak RF pulse ($\Omega_{\text{RF}} \ll A_{\parallel}$) resonant with the $|\bar{1}\rangle_e|0\rangle_a \rightarrow |\bar{1}\rangle_e|1\rangle_a$ transition results in a controlled-rotation operation, with the sensor as the control and the memory as the target, *i.e.*, a $C_e\text{ROT}_a$ gate, described by

$$U_{C_e\text{ROT}_a}^{\phi_{\text{RF}}}(\theta_{\text{RF}}) = |0\rangle\langle 0|_e \otimes \mathbb{1} + |\bar{1}\rangle\langle \bar{1}|_e \otimes e^{-i\frac{\theta_{\text{RF}}}{2}\sigma_{\phi_{\text{RF}}}}, \quad (29)$$

where θ_{RF} and ϕ_{RF} are defined analogously to the previous case. Again, for the particular case $\theta_{\text{RF}} = \pi$, Eq. (29) describes a $C_e\text{NOT}_a$ -gate:

$$U_{C_e\text{NOT}_a} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & -i \\ 0 & 0 & -i & 0 \end{pmatrix}, \quad (30)$$

where, for simplicity, $\phi_{\text{RF}} = 0$.

The alternating concatenation of three $C_e\text{NOT}_a/C_a\text{NOT}_e$ gates results in a SWAP gate:

$$U_{\text{SWAP}} = U_{C_e\text{NOT}_a} U_{C_a\text{NOT}_e} U_{C_e\text{NOT}_a} = U_{C_a\text{NOT}_e} U_{C_e\text{NOT}_a} U_{C_a\text{NOT}_e} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix}. \quad (31)$$

Consider the initial two qubit state $\rho_0 = \rho_e \otimes \rho_a$, where ρ_e takes an arbitrary form while $\rho_a = |0\rangle\langle 0|_a$:

$$\rho_0 = \begin{pmatrix} \rho_{00}^e & \rho_{01}^e \\ \rho_{10}^e & \rho_{11}^e \end{pmatrix} \otimes |0\rangle\langle 0|_a = \rho_{00}^e |00\rangle\langle 00| + \rho_{01}^e |00\rangle\langle \bar{1}0| + \rho_{10}^e |\bar{1}0\rangle\langle 00| + \rho_{11}^e |\bar{1}0\rangle\langle \bar{1}0|, \quad (32)$$

where, for the sake of clarity, we write, for instance, $|00\rangle\langle 00| = |0\rangle\langle 0|_e \otimes |0\rangle\langle 0|_a$. Then, under the application of a SWAP gate ρ_0 is transformed as follows:

$$\rho_1 = U_{\text{SWAP}} \rho_0 U_{\text{SWAP}}^\dagger = \rho_{00}^e |00\rangle\langle 00| - \rho_{01}^e |00\rangle\langle 01| - \rho_{10}^e |01\rangle\langle 00| + \rho_{11}^e |01\rangle\langle 01|, \quad (33)$$

which may be rewritten as

$$\rho_0 = |0\rangle\langle 0|_e \otimes \begin{pmatrix} \rho_{00}^e & -\rho_{01}^e \\ -\rho_{10}^e & \rho_{11}^e \end{pmatrix}. \quad (34)$$

Notice that the coherences acquire an additional negative sign which, however, is not relevant for our purposes as after applying a second SWAP gate, we recover the initial state ρ_0 .

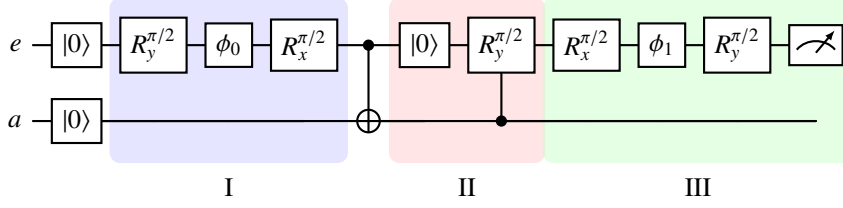


Fig. 3 Modified protocol for T_1^a -limited magnetometry. Only the population information is encoded in the nuclear memory during stage II.

Alternatively, in the case we are considering, where the memory is initialized in the $|0\rangle_a$ state, we can effectively generate two-gate SWAP operations. For encoding into the memory, we find that

$$U_{C_a \text{NOT}_e} U_{C_e \text{NOT}_a} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -i \\ 0 & -i & 0 & 0 \end{pmatrix} \quad (35)$$

produces the same state as in Eq. (33), and then

$$U_{C_e \text{NOT}_a} U_{C_a \text{NOT}_e} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & -i \\ 0 & -1 & 0 & 0 \\ 0 & 0 & -i & 0 \end{pmatrix} \quad (36)$$

recovers the initial state.

2.4 Modified sequence for T_1 -limited gradiometry

The protocol in Fig. 2 can be modified such that stage II is limited by the nuclear relaxation time T_1^a (see Fig. 3), which is typically several orders of magnitude longer than the dephasing time T_2^a . Here, the SWAP gate at the beginning of stage II is replaced by a $C_e \text{NOT}_a$ gate, hence only encoding the population information in the nuclear memory. The state of the system reads

$$\rho = \frac{1}{2} \begin{pmatrix} 1 + \sin \phi_0 & 0 \\ 0 & 1 - \sin \phi_0 \end{pmatrix} \otimes \frac{1}{2} \begin{pmatrix} 1 + \sin \phi_0 & 0 \\ 0 & 1 - \sin \phi_0 \end{pmatrix}. \quad (37)$$

Next, the sensor is reinitialized into the $|0\rangle_e$ state and a $C_a \text{ROT}_e^y(\pi/2)$ gate is applied to recover the state from the memory, such that

$$\rho = \frac{1}{4} \begin{pmatrix} 3 + \sin \phi_0 & 1 - \sin \phi_0 \\ 1 - \sin \phi_0 & 1 - \sin \phi_0 \end{pmatrix} \otimes \frac{1}{2} \begin{pmatrix} 1 + \sin \phi_0 & 0 \\ 0 & 1 - \sin \phi_0 \end{pmatrix}. \quad (38)$$

Finally, after stage III, the expected value of σ_z reads

$$\langle \sigma_z \rangle = \text{Tr}(\rho_{III} \sigma_z) = -\frac{1}{2}(\cos \phi_1 + \sin \phi_1 - \sin \phi_0 \cos \phi_1 + \sin \phi_0 \sin \phi_1), \quad (39)$$

which, assuming small ϕ_i can be approximated as

$$\langle \sigma_z \rangle \approx -\frac{1}{2} [1 + (\phi_1 - \phi_0)]. \quad (40)$$

Because only the population information is encoded into the nuclear memory, no phase is accumulated during stage II, allowing longer signal evolution times, limited only by the nuclear relaxation time.

3 Detailed description of the oscillating NV

Inducing an effective AC signal with a high frequency such as $\nu/(2\pi) = 1$ MHz is not achievable via mechanical rotation. Thus, we propose inducing small oscillations of the sensor, which allows much larger frequencies. Consider a static field the laboratory frame, $\mathbf{B} = (B_x, B_y, B_z)$. We can write the field in the oscillating frame, *i.e.*, the frame of the NV, as

$$\mathbf{B}^{\text{NV}}(t) = R_{\hat{k}}(\theta_{\text{NV}}(t))\mathbf{B}, \quad (41)$$

where $\hat{k} = (k_x, k_y, k_z)$ is a unit vector which defines the axis of oscillation and $\theta_{\text{NV}}(t)$ is the angle between the z and z^{NV} axes. This is given by $\theta_{\text{NV}}(t) = \theta_0 + \alpha \sin \nu t$, with θ_0 the initial angle between both axes, and α and ν the amplitude and frequency of the oscillations. The general rotation matrix is given by

$$R_{\hat{k}}(\theta) = \begin{pmatrix} k_x^2(1 - \cos \theta) + \cos \theta & k_x k_y(1 - \cos \theta) - k_z \sin \theta & k_x k_z(1 - \cos \theta) + k_y \sin \theta \\ k_x k_y(1 - \cos \theta) + k_z \sin \theta & k_y^2(1 - \cos \theta) + \cos \theta & k_y k_z(1 - \cos \theta) - k_x \sin \theta \\ k_x k_z(1 - \cos \theta) - k_y \sin \theta & k_y k_z(1 - \cos \theta) + k_x \sin \theta & k_z^2(1 - \cos \theta) + \cos \theta \end{pmatrix}, \quad (42)$$

such that the component of the field that is sensed by the NV center (in the reference frame of the sensor) reads

$$\begin{aligned} B_z^{\text{NV}}(t) &= [k_x k_z(1 - \cos \theta_{\text{NV}}(t)) - k_y \sin \theta_{\text{NV}}(t)]B_x \\ &+ [k_y k_z(1 - \cos \theta_{\text{NV}}(t)) + k_x \sin \theta_{\text{NV}}(t)]B_y \\ &+ [k_z^2(1 - \cos \theta_{\text{NV}}(t)) + \cos \theta_{\text{NV}}(t)]B_z. \end{aligned} \quad (43)$$

Because we assume a small oscillation amplitude α , we can approximate

$$\begin{aligned} \sin \theta_{\text{NV}}(t) &\approx \sin \theta_0 + \alpha \cos \theta_0 \sin \nu t, \\ \cos \theta_{\text{NV}}(t) &\approx \cos \theta_0 - \alpha \sin \theta_0 \sin \nu t, \end{aligned} \quad (44)$$

and knowing that static terms will have no effect as we will apply dynamical decoupling, we find

$$\begin{aligned} B_z^{\text{NV}}(t) &\approx \alpha \sin \nu t [k_x k_z \sin \theta_0 - k_y \cos \theta_0]B_x \\ &+ \alpha \sin \nu t [k_y k_z \sin \theta_0 + k_x \cos \theta_0]B_y \\ &+ \alpha \sin \theta_0 \sin \nu t (k_z^2 - 1)B_z. \end{aligned} \quad (45)$$

Now, by setting $\theta_0 = 0$,

$$B_z^{\text{NV}}(t) = \alpha \sin \nu t (k_x B_y - k_y B_x), \quad (46)$$

such that $B_z^{\text{NV}}(t) = \alpha \sin \nu t B_y$ for $k_x = 1$ and $k_y = 0$, and $B_z^{\text{NV}}(t) = -\alpha \sin \nu t B_x$ for $k_x = 0$ and $k_y = 1$. Similarly, setting $\theta_0 = \pi/2$,

$$B_z^{\text{NV}}(t) = \alpha \sin \nu t (k_x k_z B_x + k_y k_z B_y) + \alpha \sin \nu t (k_z^2 - 1) B_z, \quad (47)$$

which is zero for $k_{x,y} = 0$ and $k_z = 1$, and $B_z^{\text{NV}}(t) = -\alpha \sin \nu t B_z$ for $\hat{k}$ in the xy plane.

The evolution of the sensor spin state due to the effective AC signal induced by the oscillation is thus described by

$$\mathcal{H}(t) = |\gamma_e| B_z^{\text{NV}}(t) S_z = \alpha |\gamma_e| \sin \nu t B_i S_z, \quad (48)$$

where $B_i = -B_x, B_y, -B_z$. To capture this signal, we apply a dynamical decoupling sequence such that the evolution operator in the toggling frame reads

$$U = \exp(-i \mathcal{H}_{\text{eff.}}(2\tau N_p)), \quad (49)$$

where $\tau = 1/(4\nu)$ is the interpulse separation, N_p the number of pulses in the sequence and

$$\mathcal{H}_{\text{eff.}} = \frac{S_z}{2\tau N_p} \int_0^{2\tau N_p} \mathcal{F}(t) \mathcal{H}(t) dt = \frac{2\alpha |\gamma_e| B_i N_p}{\nu} S_z. \quad (50)$$

Here, $\mathcal{F}(t)$ is an odd modulation function imprinted by the dynamical decoupling sequence, which is unity at $t = 0$ and changes sign when every time a π -pulse is applied. Then, the phase accumulated by the sensor is

$$\phi = \frac{2\alpha N_p |\gamma_e| B_i}{\nu}. \quad (51)$$

4 Modelling of magnetic anomalies

In order to model magnetic anomalies, we will assume that the measurement occurs in the far-field regime, such that the source of the anomaly, which is considered to be a sphere, can be approximated as a dipole:

$$\mathbf{B}_{\text{an.}}(\mathbf{r}) = \frac{\mu_0}{4\pi} \left(\frac{3(\mathbf{m} \cdot \mathbf{r})\mathbf{r}}{r^5} - \frac{\mathbf{m}}{r^3} \right), \quad (52)$$

where μ_0 is the magnetic susceptibility of vacuum and $\mathbf{m}$ is the magnetic dipole moment, which is related to the magnetization $\mathbf{M}$ of the source by $\mathbf{M} = \mathbf{m}/V$, with V its volume. This magnetization is due to an external or inducing field $\mathbf{H}_{\text{ext.}}$ (in this case, this is the geomagnetic field) and an internal field $\mathbf{H}_{\text{int.}}$ which originates from the induced magnetization, such that

$$\mathbf{M} = \chi \mathbf{H} = \chi (\mathbf{H}_{\text{ext.}} + \mathbf{H}_{\text{int.}}), \quad (53)$$

with χ the magnetic susceptibility of the source. In this case where the inducing field is the geomagnetic field, we have that $\mathbf{H}_{\text{ext.}} = \mathbf{B}_{\text{gm}}/\mu_0$. The internal field, on the other hand, is given by the following expression:

$$\mathbf{H}_{\text{int.}}(\mathbf{r}) = \frac{1}{4\pi} \int_{V'} \mathbf{M}(\mathbf{r}') \nabla^2 \frac{1}{|\mathbf{r} - \mathbf{r}'|} d^3 \mathbf{r}'. \quad (54)$$

For a uniformly magnetized sphere, it can be easily shown that

$$\mathbf{M} = \frac{3\chi}{3 + \chi} \mathbf{H}_{\text{ext.}}, \quad (55)$$

which in the limit of high susceptibility (*e.g.*, for a ferromagnetic source), $\mathbf{M} \rightarrow 3\mathbf{H}_{\text{ext.}}$. Then, the magnetic field emitted by the source, *i.e.*, the magnetic anomaly, can be described by

$$\mathbf{B}(\mathbf{r}) = \frac{V}{4\pi r^3} \frac{3\chi}{3 + \chi} [3(\mathbf{H}_{\text{ext.}} \cdot \hat{\mathbf{r}})\hat{\mathbf{r}} - \mathbf{H}_{\text{ext.}}]. \quad (56)$$

The figure shows the magnetic field emitted by a spherical ferromagnetic source of radius $R = 16$ cm, calculated using Eq. (56) over a square of length $2L = 10$ m at a height $h = 1$ m.

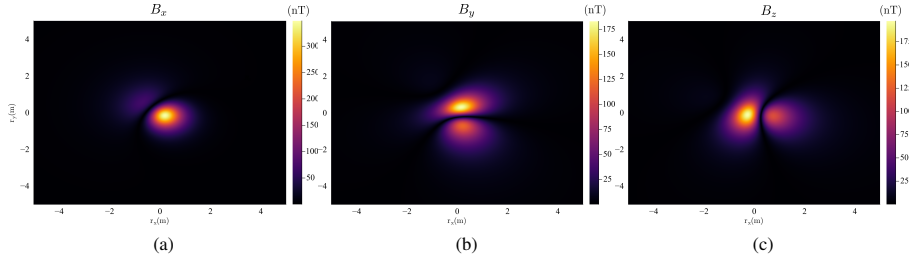


Fig. 4 Magnetic field emitted by a ferromagnetic sphere of radius $R = 16$ cm in the far-field regime. The colour plot shows the intensity of the anomaly field (nT) over a square of side $2L = 10$ m at a height $h = 1$ m. The geomagnetic field that induces the magnetization of the sphere has an intensity $B = 45.85 \mu\text{T}$, a colatitude $\theta = 105.63^\circ$ and a longitude $\phi = 320.72^\circ$.

5 Modeling of magnetic fields from neuron action potentials

Neuron action potentials (APs) consist in the depolarization and repolarization of a neuron under a stimulus, which is due to the movement of ions in and out the cell. This increase in the intracellular voltage Φ of the neuron results in an electric impulse which is transferred to another neuron through an axon, which we model as a wire, such that the current density through the axon is given by Ohm's law:

$$\mathbf{J} = -\sigma \nabla \Phi(z, \rho, t), \quad (57)$$

where σ is the conductivity of the axon and ρ is the radial coordinate in cylindrical coordinates. Choosing the direction of propagation of the impulse to be aligned with the z direction, we find the current intensity to be

$$I = -\pi\sigma r_a^2 \frac{\partial\Phi(z, \rho, t)}{\partial z}, \quad (58)$$

with r_a the radius of the axon. Then, assuming a constant conduction velocity,

$$\frac{\partial\Phi(z, \rho, t)}{\partial t} = -v_c \frac{\partial\Phi(z, \rho, t)}{\partial z}. \quad (59)$$

Finally, using Biot-Savart law, we find the magnetic field associated to the electrical impulse,

$$B_x(t) = \frac{\mu_0 I}{2\pi\rho} = \frac{\mu_0\sigma r_a^2}{2v_c\rho} \frac{\partial\Phi(z, \rho, t)}{\partial t}, \quad (60)$$

where $s \equiv \frac{\mu_0\sigma r_a^2}{2v_c\rho}$ is a proportionality constant. Following [1], we take $s = 7.6\text{pT}/(\text{V/s})$. Note that we consider the direction of the field at the position of the sensor to be perpendicular to the propagation of the electrical impulse, which we take to be aligned with the x axis. Then, ρ is the distance to the sensor. In this work, we model the intracellular voltage to replicate the experimental results of [1].

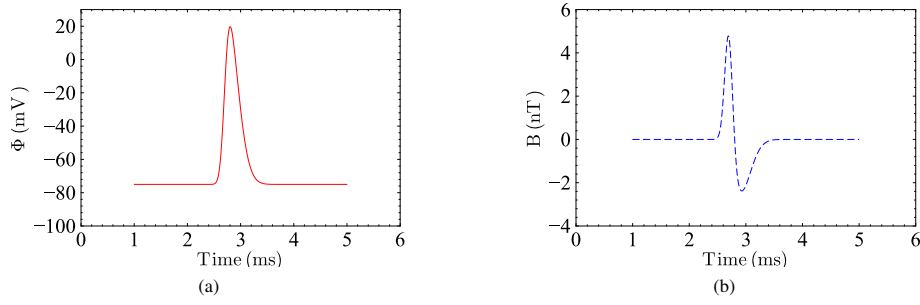


Fig. 5 Gradiometry for magnetophysiology: neuron action potential. (a) Intracellular voltage Φ that originates the electric impulse. (b) The magnetic field calculated from the intracellular voltage.

References

- [1] Barry, J.F., Turner, M.J., Schloss, J.M., Glenn, D.R., Song, Y., Lukin, M.D., Park, H., Walsworth, R.L.: Optical magnetic detection of single-neuron action potentials using quantum defects in diamond. *Proceedings of the National Academy of Sciences* **113**(49), 14133–14138 (2016) <https://doi.org/10.1073/pnas.1601513113>