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PAPER

Quantum sensing of electric field distributions of liquid electrolytes with NV-centers in nanodiamonds

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E-mail: roland.nagy@fau.de**Keywords:** NV-Center, quantum sensing, nano diamonds, battery cells, electrolyteSupplementary material for this article is available [online](#)

Abstract

To use batteries as large-scale energy storage systems it is necessary to measure and understand their degradation *in-situ* and *in-operando*. As a battery's degradation is often the result of molecular processes inside the electrolyte, a sensing platform which allows to measure the ions with a high spatial resolution is needed. Primary candidates for such a platform are NV-centers in diamonds. We propose to use a single NV-center to deduce the electric field distribution generated by the ions inside the electrolyte through microwave pulse sequences. We show that the electric field can be reconstructed with great accuracy by using a protocol which includes different variations of the free induction decay to obtain the mean electric field components and a pulse sequence consisting of three polarized π -pulses to measure the electric field's standard deviation σ_E . From a semi-analytical ansatz we find that for a lithium ion battery there is a direct relationship between σ_E and the ionic concentration. Our results show that it is therefore possible to use NV-centers as sensors to measure both the electric field distribution and the local ionic concentration inside electrolytes.

1. Introduction

Rechargeable batteries play an important role for our society and are a key ingredient for the transition towards renewable energy sources [1–3]. As the production of batteries is accompanied with a considerable use of resources, recyclable [4] batteries with a long lifetime are needed. The latter is limited by degradation mechanisms, such as the formation of solid-electrolyte interfaces [5] or lithium-plating [6] which can reduce the battery's capacity with increasing cell age [7]. As these processes happen on a molecular level [5], a sensor which is capable of monitoring the ionic concentration *in-situ* and *in-operando* with high spatial and temporal resolutions is needed. Even though MRI allows to reconstruct transport properties [8, 9] of a battery by measuring the macroscopic concentration gradients which form across the electrochemical cell, it has a limited spatial resolution [8]. Tools which allow to perform measurements inside the electrochemical cell with a better resolution are still absent [5].

It has been demonstrated that the nitrogen-vacancy (NV) center in diamond (see figure 1(a)) is a high-resolution quantum sensor, which can detect oscillating or fluctuating [10–13] magnetic fields with nano- [14, 15] and even subpico-Tesla [16] sensitivities. Besides this, NV-centers have great ability for the detection of electric fields. They can not only detect DC [17, 18] or AC [19] electric fields with remarkable precision but are additionally capable of detecting the change in electric field generated by a single fundamental charge within the diamond lattice [20] as well as their spatial location [21]. This electric field

sensitivity was used by [22] to show that, based on theoretical considerations, bulk NV-centers can work as electrochemical sensors if they are in contact with an electrolyte solution.

Here we show that nanodiamonds equipped with single NV-centers can act as *in-situ* electric field sensors inside liquid electrolytes (figure 1(a)). We assume that after being inserted into the electrolyte, the nanodiamond will find an equilibrium position near the electrodes and inside the separator of the battery. If the nanodiamond does not stay static during the experiments it will be necessary to track its position and orientation in real time (see supplementary information). Due to the charge of the electrodes, the electrolyte will be under the influence of an electric field [23]. During (dis)charge, the ionic concentration profile changes [24], which is accompanied by a change of the electric field inside the electrolyte [24]. Measurements of the mean electric field values inside the liquid electrolyte therefore allow insights into a battery's concentration profile, which can then be used to infer its state of charge [25]. By exploiting how transverse and axial electric fields act on the NV-center's ground state spin states (figure 2(a)), we find variations of the free-induction decay (FID) pulse sequence, which allow to measure these mean electric field components. Fluctuations in the ionic concentration, e.g. because of diffusion, will cause fluctuations in the electric field [22] (cf figure 1(b)). In the following, we characterize these fluctuations as deviations σ_E and show that it is possible to use a pulse sequence composed of three circularly polarized π -pulses to additionally obtain their value if the mean electric field is known. From a semi-analytical ansatz we demonstrate exemplarily for a lithium ion battery (LIB) that there is a direct relationship between the electric field's standard deviation and the local ionic concentration. A nanodiamond with a single NV-center can therefore work as a sensor which allows to simultaneously reconstruct the electric field distribution and to measure the ionic concentration with high spatial resolution.

2. Electric field distribution in liquid electrolytes

Before introducing measurements of the electric field distribution by the NV-center, we would like to develop an analytic expression of the electric field induced inside the nanodiamond by the positive and negative ions of the electrolyte.

The potential Φ at position $\mathbf{r}$ inside the nanodiamond due to a single charge q at position $\mathbf{b}$, is described by Poisson's equation

$$\nabla^2 \Phi(\mathbf{r}) = -\frac{\rho(\mathbf{r})}{\epsilon}. \quad (1)$$

Here $\epsilon = \epsilon_0 \epsilon_i$ with $i = e, \text{ND}$, are the permittivities of, respectively, the electrolyte and the nanodiamond in terms of the vacuum permittivity ϵ_0 and ρ is the charge density induced by q . The solution inside the nanodiamond, Φ_{ND} (see supplementary information for the detailed derivation), allows to obtain the electric field at the center of the nanodiamond, which is

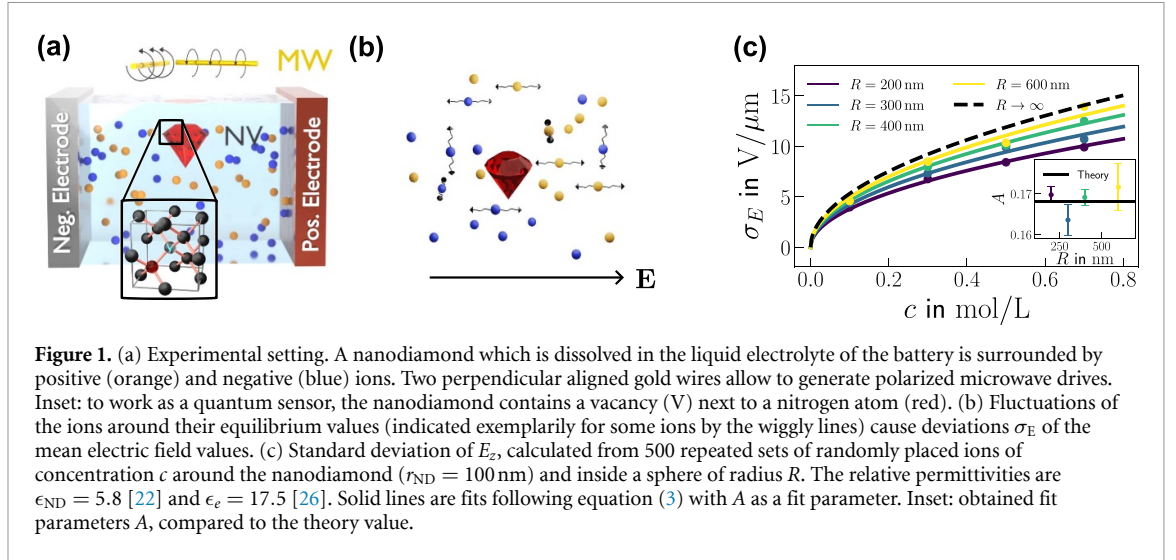
$$\mathbf{E}^{\text{ND}} = \frac{q}{4\pi\epsilon_0} \frac{3}{2\epsilon_e + \epsilon_{\text{ND}}} \frac{\mathbf{b}}{b^3}. \quad (2)$$

By considering the positions of ions of a molar concentration c to be uniformly distributed within a sphere of radius R around a nanodiamond (radius r_{ND}), the standard deviation of the electric field distribution at the center of the nanodiamond is

$$\sigma_{E_z} = A \sqrt{c \left(\frac{1}{r_{\text{ND}}} - \frac{1}{R} \right)} \quad (3)$$

$$A = \frac{|q|}{\epsilon_0 (2\epsilon_e + \epsilon_{\text{ND}})} \sqrt{\frac{3N_A}{4\pi}}.$$

Here N_A is Avogadro's constant. To validate equation (3), we simulated the standard deviation of 500 sets of uniformly and randomly placed ions for different molar ionic concentrations (see figure 1(c)). As it is the most widely used electrolyte of LIBs [27], we chose LiPF_6^- with $\epsilon_e = 17.5$ [26]. The total electric field was calculated as the linear sum of equation (2) for all randomly placed ions around a nanodiamond with $r_{\text{ND}} = 100 \text{ nm}$ [28]. As it can be seen from figure 1(c), the expected A value is in fair agreement with the simulations. From equation (3) it can be calculated that for a nanodiamond with $r_{\text{ND}} = 100 \text{ nm}$, 90% of the electric field fluctuations (compared to $\sigma_{E_z}(R = \infty)$ as following from equation (3)) are due to ions within a sphere of $R = 1 \mu\text{m}$ around the nanodiamond. This implies that electric field fluctuations only affect the nanodiamond within micrometer range and the system is limited by the confocal volume of the experimental setup, which typically is $\sim 1 \mu\text{m}^3$ [29, 30].



3. Sensing of static electric fields inside electrolytes

As illustrated in figure 1(b), the nanodiamond will be subject to a static electric field, whereas fluctuations of the ions will cause deviations σ_E from these mean values. Before we introduce measurement techniques of these perturbations in section 4, we will first show how the mean electric field values can be sensed through variations of the FID.

In the following we assume that the nanodiamond's position and orientation stays static on the timescales at which the pulse sequences are performed. As discussed in the supplementary information, it is possible to track the nanodiamond's position and orientation in real-time.

An electric field $\mathbf{E}$ can in cylindrical coordinates be expressed by its axial component E_z , its transverse projection $E_\perp = \sqrt{E_x^2 + E_y^2}$ and an angle ϕ_E , which defines the projections onto the x and y axis as $E_x = E_\perp \cos \phi_E$ and $E_y = E_\perp \sin \phi_E$. The total Hamiltonian which describes the NV-center in presence of electric and axial magnetic fields will in the following be denoted as $\hat{\mathcal{H}}_0$. By taking into account that the NV-center can be driven by two perpendicular microwave wires (see figure 1(a)) with amplitude Ω , frequency ω_d and a phase ϕ_{MW} between each other, the total ground state Hamiltonian in a frame rotating with ω_d is $\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_d$ (see supplementary information), where

$$\begin{aligned}\hat{\mathcal{H}}_0 &= (\Delta + \xi_z) \hat{S}_z^2 + \beta_z \hat{S}_z - \frac{\xi_\perp}{2} \left(\hat{S}_+^2 e^{i\phi_E} + h.c. \right) \\ \hat{\mathcal{H}}_d &= \frac{\Omega}{\sqrt{2}} (\eta_+ \sigma_+ + \eta_- \sigma_- + h.c.) .\end{aligned}\quad (4)$$

Here $\Delta = D - \omega_d$ is the detuning between the zero-field splitting, $D = 2.87$ GHz [31], and the microwave drive frequency. S_i , $i = x, y, z$, are the spin-1 operators, which can be used to define ladder operators $S_\pm = S_x \pm i S_y$. $\sigma_\pm = |\pm 1\rangle\langle 0|$ are operators which describe transitions between $|0\rangle$ and, respectively, $|\pm 1\rangle$. Frequency contributions generated by electric fields $\mathbf{E}$ as well as strain $\mathbf{\Pi}$ within the diamond lattice are considered through $\xi_\perp = d_\perp (E_\perp + \Pi_\perp)$ and $\xi_z = d_z (E_z + \Pi_z)$ with $d_z = 0.35$ Hz cm V⁻¹ and $d_\perp = 17$ Hz cm V⁻¹ [32]. The constant offset in the total electric field contributions $\xi_\perp$ and ξ_z as generated by the strain is different for each individual nanodiamond and can, for instance, be determined through ODMR (optically detected magnetic resonance) measurements [33]. $\beta_z = \gamma_e B_z$ with $\gamma_e = 28$ GHz T⁻¹ [34] describes contributions due to axial magnetic fields.

The phase factors $\eta_\pm = (1 \mp i e^{-i\phi_{MW}})/2$ which enter into equation (4), allow to describe the transitions which are caused by circularly ($\phi_{MW} = \pm\pi/2$) or linearly ($\phi_{MW} = 0$) polarized microwave drives [35]. We assume the linewidth of the drive to be broad enough such that hyperfine interaction, for instance with the nitrogen nucleus, can be neglected [36]. The time-evolution operators of $\hat{\mathcal{H}}_d$, $\hat{R}(t) = e^{-i\hat{\mathcal{H}}_d t}$ (see supplementary information), show that one can induce Rabi oscillations between $|0\rangle$ and $|1\rangle$ for right circularly polarized drives and $|0\rangle \leftrightarrow |-1\rangle$ for left circular polarizations. Linearly polarized drives allow to drive transitions between $|0\rangle$ and both $|\pm 1\rangle$.

In absence of microwave drives, the $|\pm 1\rangle$ states are symmetrically mixed by $\xi_\perp$ and axial electric fields effectively shift $|0\rangle$ from $|\pm 1\rangle$, which can be seen from $\hat{F}(\tau) = e^{-i\hat{\mathcal{H}}_0 \tau}$ (see supplementary information) and

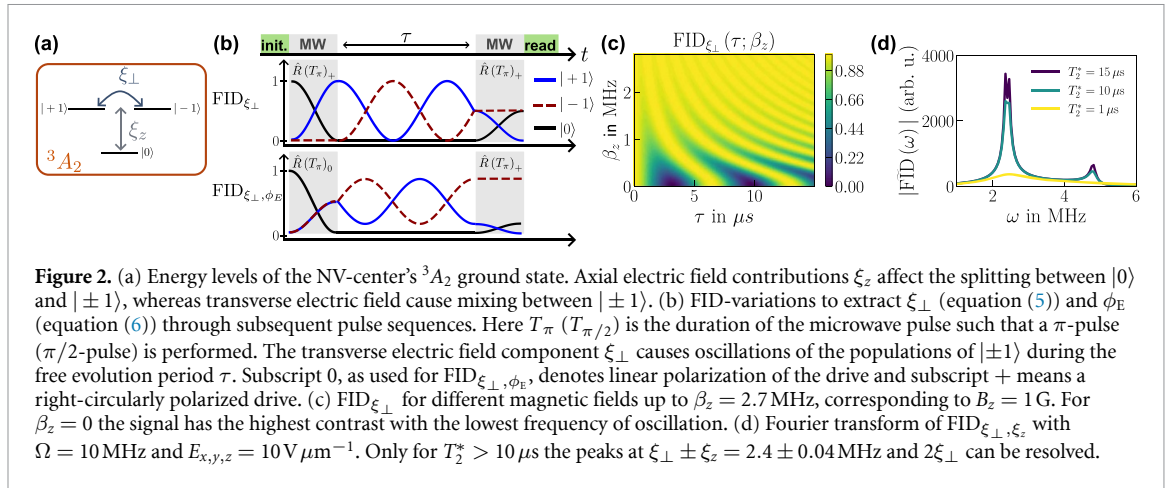


figure 2(a). As axial and transverse electric fields thus act differently on the $|m_s = 0, \pm 1\rangle$ states of the NV-center, one can derive variations of the FID, which allow to extract these electric field components.

3.1. Measurement of electric field components

The FID consists of two microwave pulses separated by a free evolution period τ . Electric field contributions $\xi_\perp$, ϕ_E and ξ_z can be sensed through FID-variations, (see figure 2(b)). The NV-center can be initialized into its $|0\rangle$ state via the excitation with green laser light, followed by intersystem-crossing [37]. This state can then be driven to $-i|1\rangle$ through a right-circularly polarized π -pulse, denoted as $\hat{R}(T_\pi)_+$, and will be influenced by both axial magnetic as well as transverse electric fields during the free evolution interval. The latter induce mixing with $|-1\rangle$, as can be seen in figure 2(a). By using a microwave π -pulse with the same polarization as the initial one, the transferred population from $|1\rangle$ to $|-1\rangle$ can be obtained, as this pulse transfers the population from $|+1\rangle$ to $|0\rangle$ (cf figure 2(b)), which will then be read out through the final pulse with green laser light. This is resembled in the FID-signal

$$\begin{aligned} \text{FID}_{\xi_\perp}(\tau) &= |\langle 0 | \hat{R}(T_\pi)_+ \hat{F}(\tau) \hat{R}(T_\pi)_+ | 0 \rangle|^2 \\ &= \cos^2 \left(\tau \sqrt{\beta_z^2 + \xi_\perp^2} \right) \\ &\quad + \frac{\beta_z^2}{\beta_z^2 + \xi_\perp^2} \sin^2 \left(\tau \sqrt{\beta_z^2 + \xi_\perp^2} \right), \end{aligned} \quad (5)$$

which is a measure of the population which has been transferred from $|1\rangle$ to $|-1\rangle$ (cf figure 2(b)). In figure 2(c) one can see this FID-signal for $E_{x,y} = 2 \text{ V } \mu\text{m}^{-1}$ as a function of the free evolution time τ for β_z values up to 2.8 MHz, which corresponds to $B_z = 1$ G. For our simulations we assume electric fields on the order of $\sim \text{V } \mu\text{m}^{-1}$ as derived in earlier studies of NV-centers in contact to liquid electrolytes [22]. Besides having a decreased contrast for $\beta_z \neq 0$, the frequency $\sqrt{\beta_z^2 + \xi_\perp^2}$ of the FID-oscillations depends on both axial magnetic and transverse electric fields. It is therefore needed to perform the measurements in a magnetically shielded environment, for example by a μ -metal as in [33, 38]. In the following it will be assumed that all measurements are performed without any magnetic field being present.

The transverse electric field components are uniquely defined through ϕ_E , as $\xi_x = \xi_\perp \cos \phi_E$ and $\xi_y = \xi_\perp \sin \phi_E$. A superposition state $-e^{i\pi/4}(|1\rangle + |-1\rangle)/\sqrt{2}$ generated through a linearly polarized π -pulse (considered via $\hat{R}(T_\pi)_0$, see supplementary information) will additionally to $\xi_\perp$ also be affected by ϕ_E as this phase differs in its sign for $|1\rangle$ and $|-1\rangle$ (see supplementary information). If either $|1\rangle$ or $|-1\rangle$ is projected to $|0\rangle$ through the final microwave pulse, one obtains an FID-signal, which both depends on $\xi_\perp$ and ϕ_E ,

$$\text{FID}_{\phi_E, \xi_\perp}(\tau) = \frac{1}{2} (1 - \sin(2\tau\xi_\perp) \sin \phi_E). \quad (6)$$

One can obtain ϕ_E as the relative fraction between the value of the FID-signal at $\tau = 0$ and its first maxima at $2\tau\xi_\perp = \pi/2$,

$$\frac{\text{FID}_{\phi_E, \xi_\perp} \left(\tau = \frac{\pi}{2} \frac{1}{2\xi_\perp} \right)}{\text{FID}_{\phi_E, \xi_\perp}(\tau = 0)} = 1 - \sin \phi_E. \quad (7)$$

By using $\text{FID}_{\xi_{\perp}}$ and $\text{FID}_{\xi_{\perp}, \phi_E}$, it is therefore possible to not only determine the electric field's transverse component, but also to obtain the projection onto the x and y axes, which are determined through ϕ_E .

Axial electric field contributions ξ_z cause a Stark shift between $|0\rangle$ and $|\pm 1\rangle$. A superposition state $(|0\rangle - i| -1\rangle)/\sqrt{2}$ generated by a circularly polarized $\pi/2$ -pulse will therefore be affected both by ξ_z and $\xi_{\perp}$. If the final microwave $\pi/2$ -pulse has the same polarization as the initial one, an FID-signal is obtained which depends both on $\xi_{\perp}$ and ξ_z ,

$$\text{FID}_{\xi_z, \xi_{\perp}}(\tau) = \frac{1}{4} (1 - 2 \cos(\tau \xi_{\perp}) \cos(\tau \xi_z) + \cos^2(\tau \xi_{\perp})) , \quad (8)$$

if the NV-center was driven with $\omega_d = D$. The Fourier transform of equation (8) (see supplementary information),

$$\widetilde{\text{FID}}(\omega > 0) = \frac{\pi}{4} \left(\frac{1}{2} \delta(2\xi_{\perp} - \omega) - \delta(\xi_{\perp} + \xi_z - \omega) - \delta(\xi_{\perp} - \xi_z - \omega) \right) , \quad (9)$$

shows, that ξ_z can be measured if it is possible to spectrally resolve $\xi_{\perp} \pm \xi_z$. To study this, we numerically [39, 40] simulated $\text{FID}_{\xi_z, \xi_{\perp}}$ and included dephasing at rates $1/T_2^*$ through a Lindblad operator $\sqrt{1/T_2^*} S_z$ for T_2^* in the range up to $15 \mu\text{s}$ (see figure 2(d)). One can resolve $\xi_{\perp} \pm \xi_z$ for nanodiamonds with $T_2^* > 10 \mu\text{s}$, which is higher than the value of typical nanodiamonds [41, 42]. For a nanodiamond with $T_2^* \approx 15 \mu\text{s}$ it would be possible to distinguish between $\xi_{\perp}$ and ξ_z and therefore to determine the projection of the electric field onto the symmetry axis of the NV-center.

4. Influence of fluctuating electric fields

It can be assumed that the ions surrounding the nanodiamond will not stay static for the timescales in which measurements are performed but will be subject to, for instance, drift and diffusion. These fluctuations will affect the electric field inside the nanodiamond. The total electric field distribution can therefore be described by a normal distribution with mean E_m and standard deviation σ_E . Due to the limited T_2^* of nanodiamonds, the FID pulse sequences as introduced before will be mainly suitable for the measurement of the average electric fields (see supplementary information). As it is shown in figure 3(a), we propose a pulse sequence composed of three polarized π pulses (TPPP), where after the first free evolution interval, a π -pulse with right-circular polarization is performed, before the spin is allowed to precess freely during a second free evolution interval τ . As this circularly polarized π -pulse only affects the $|0\rangle$ and $|+1\rangle$ states, the effect of the transverse electric fields is not cancelled. Before being read out, a right-circularly polarized π -pulse is applied, which leads to a signal $\text{TPPP}(\tau) = (1 - \cos(2\tau \xi_{\perp}))^2 / 4$. Simulations of this pulse sequence show that the averages (see supplementary information for an example) can be fit by

$$\langle \text{TPPP}(\tau) \rangle = \frac{1}{4} \left[1 - \cos(2\tau \xi_{\perp}) e^{-\tau/T_2} \right]^2 . \quad (10)$$

Here T_2 is the sum of the intrinsic spin coherence time $T_{2,\text{int.}}$ and a contribution due to the fluctuating electric fields,

$$\frac{1}{T_2} = \frac{1}{T_{2,\text{int.}}} + \frac{1}{T_{2,E}} . \quad (11)$$

In figure 3(b), one can see T_2 as a function of the electric field's standard deviation σ_E , where solid lines are $T_{2,E} = \alpha E_m / \sigma_E^2$ in terms of a fit parameters α . The total spin coherence time is therefore strongly affected by σ_E and the mean electric field value $E_m = E_x, E_y, E_z$. To allow for the sensing of σ_E through the TPPP pulse sequence, the contribution onto the total dephasing of the spin must be dominated by the electric field fluctuations, i.e. $1/T_{2,E} > 1/T_{2,\text{int.}}$. For the simulations shown in figure 3 we have chosen $T_{2,\text{int.}} = 100 \mu\text{s}$ as reported for Hahn-echoes with nanodiamonds [28]. Based on simulations of cluster correlation expansion methods (see supplementary information) this is a reasonable choice for the TPPP pulse sequence if spin impurities within the diamond lattice are identified as the main source of decoherence. If the mean electric field is strong enough, it is also possible to measure the decay of TPPP signal induced by the electric field fluctuations for $T_{2,\text{int.}} < 100 \mu\text{s}$ (see supplementary information for an example with $T_{2,\text{int.}} = 30 \mu\text{s}$). If the mean transverse electric field has been sensed by the FID sequence as shown in equation (5), it is therefore possible to derive the electric field's standard deviation, which together with $\xi_{\perp}$, ϕ_E and ξ_z defines the electric field distribution. As there is a direct relationship between σ_E and the local ionic concentration (see figure 1(c)), the proposed TPPP pulse sequence additionally allows to use the NV-center inside the nanodiamond as a local concentration sensor.

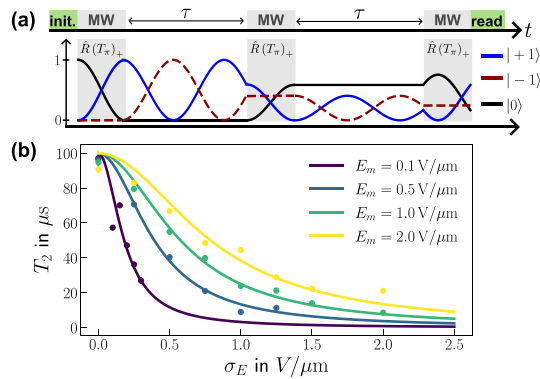


Figure 3. (a) TPPP pulse sequence used to simulate equation (10) including the time evolution of the states during the microwave pulses and the free evolution intervals. (b) Total T_2 for numerically [39, 40] simulated TPPP with $T_{2,int} = 100 \mu\text{s}$. At every timestep at which the time-evolution of the NV-center is simulated, the electric field components are sampled from a normal distribution with mean E_m and standard deviation σ_E (see supplementary information). For the simulations a drive of $\Omega = 10 \text{ MHz}$ was used. Solid lines are fits of $\alpha E_m / \sigma_E^2$. Every trajectory was obtained from 1000 individual simulations. Error bars of one standard deviation are smaller than the data points.

5. Conclusion and outlook

In conclusion we have shown here a full reconstruction of the mean electric field generated in a liquid electrolyte, through the spin control of a quantum sensor immersed in the electrolyte. We have found exact expressions correlating the electric field components with the FID of the sensor spin, and the dependence of the variance on the spin-echo measurements. In summary, we were able to deduce the electric field distribution and propose a protocol, which allows to characterize the battery's state of charge and to obtain insight into the transport processes happening inside the electrolyte, like for instance diffusion, which are characterized by σ_E . We envisage that with improved modeling of the electric field distribution in liquid electrolytes we can additionally explore the ability of nanodiamonds equipped with NV-centers for the detection of electric field inhomogeneities within the battery. Combined with better quantum control methods, for example using correlation spectroscopy [43] or combinations of different dynamical decoupling protocols [44], we could enhance the sensitivity of the sensor to the local electric-field environment, allowing for the full *in-situ* monitoring of the battery using the liquid electrolyte.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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