

Mapping oscillating magnetic fields around rechargeable batteries

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Abstract

Power storage devices such as batteries are a crucial part of modern technology. The development and use of batteries has accelerated in the past decades, yet there are only a few techniques that allow gathering vital information from battery cells in a noninvasive fashion. A widely used technique to investigate batteries is electrical impedance spectroscopy (EIS), which provides information on how the impedance of a cell changes as a function of the frequency of applied alternating currents. Building on recent developments of inside-out MRI (ioMRI), a technique is presented here which produces spatially-resolved maps of the oscillating magnetic fields originating from the alternating electrical currents distributed within a cell. The technique works by using an MRI pulse sequence synchronized with a gated alternating current applied to the cell terminals. The approach is benchmarked with a current-carrying wire coil, and demonstrated with commercial and prototype lithium-ion cells. Marked changes in the fields are observed for different cell types.

Keywords: Current imaging, Alternating current, Oscillating field, Triggered acquisition, Rechargeable batteries

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1 Introduction

2 Power storage devices form the basis for portable electronics technology
3 as well as for the development of electric transportation options. The demand
4 for rechargeable batteries will significantly increase in the coming years, yet
5 technology for adequate assessment of commercial-type cells is currently rel-
6 atively limited. In particular, it is vital to establish detailed noninvasive
7 diagnostics techniques that would allow identifying defects, predicting life-
8 times, and determining critical failure mode progressions.

9 A commonly used technique to investigate batteries is electrical impedance
10 spectroscopy (EIS) [1]. EIS is based on applying alternating current (AC),
11 and recording the complex impedance as a function of current frequency
12 [2]. EIS is further widely used to characterize materials as well as biological
13 tissues based on the frequency response of their electrical conductivity and
14 permittivity [3, 4, 5]. When used with batteries, EIS is often employed to
15 assess information about internal impedance as a function of temperature,
16 depth of discharge, and at different points in the cycling process [6, 7, 8].
17 Changes in the frequency response of the internal impedance of an electro-
18 chemical energy storage device provide insights into cell performance, includ-
19 ing, for example, measures of the uniformity of the material distribution, of
20 the quality of solid electrolyte interphase (SEI), of the electrode porosity, and
21 of adsorption kinetics [9, 10, 11]. A major limitation of EIS is that it records
22 only the global values for the device, and no spatial resolution is provided.
23 While there exist techniques such as Localized Electrochemical Impedance
24 Spectroscopy [12] or scanning electrochemical microscopy (AC-SECM)[13],
25 these methods require access to the electrode surface and direct electrical
26 contact, which is problematic for realistic or commercial-type cells and many
27 other devices.

28 While conventional MRI is not able to investigate the processes within
29 commercial batteries due to the difficulty for radiofrequency (RF) to pene-
30 trate through their metal casings or through the electrode layers, the inside-
31 out MRI (ioMRI) approach is able to reveal important information by ana-
32 lyzing the magnetic susceptibility changes of the cell's materials via the mea-
33 surement of the magnetic field distributions around the cell. This approach
34 has been employed to detect susceptibility variations caused by changes in
35 the cell as well as to investigate changes in direct current distributions across
36 the device [14, 15, 16, 17, 18].

37 In addition, other indirect parameters such as the spatial distribution of

the magnetic field generated outside the battery caused by the current can be used as a measure of internal battery properties.

A number of MRI methods have been developed to detect electric currents via measurements of the corresponding magnetic fields using synchronized pulse sequence elements [19, 20, 21, 22, 23, 24, 25]. An approach based on a tuned spin-lock field exploits the coherent interaction between the spin-lock pulse and a fluctuating magnetic field produced by electrical currents [19]. While this approach is potentially quite powerful, there are a number of practical difficulties which limit its successful implementation for our intended application. In particular, this approach exhibits problems at low frequencies due to RF imperfections and $T_{1\rho}$ relaxation. Other techniques comprise current density imaging (CDI) sequences[21, 22, 23] and steady state free precession (SSFP) approaches[20], both based on exploiting phase changes in the magnetic resonance signals caused by current-generated local magnetic fields. However, both approaches require precise synchronization of pulse sequence elements with the AC frequency.

Yang *et al.* showed, that a gradient echo sequence with a sufficient Field-of-View exhibits ghost images at regular distances which depend on the AC frequency of the current passing through a wire at a specific orientation [25]. The Fourier transform of the signal revealed these images. More recently, a method based on multiple repetition times to extend the range of frequencies accessible in measurements of brain waves was proposed by Kim *et al.* [24]. A Fourier transform of a series of images with a fixed repetition time was employed to recover a spectrum of oscillating field frequencies. With this approach, the delay between a certain point of the AC wave and the start of the acquisition is fixed, yet unknown. In this approach, the spectral width of the encoding of the oscillating field is not known. A second experiment set acquired with a different repetition time can recover accurate frequency information without this *a priori* information.

Building on lessons learnt from prior work, we present here an improved technique, suitable for mapping the oscillating magnetic fields around battery cells. In, particular the use of triggers from the current source to spectrometer as well as gating the AC current allow for more control of the experiment. This sequence and method of potentially performing battery diagnostics is henceforth referred to as inside-out Triggered Alternating Current ImagiNg employing Gating (io-TRACING). In contrast to CDI-based techniques, it is possible to scan a range of frequencies with one measurement instead of having to tailor all pulse sequence element delays to a specific

frequency. Furthermore, it is possible to obtain images at very low frequencies, e.g. **0.1-1 Hz** **0.1 - 10 Hz**, which is a particularly useful range for devices such as batteries. **In this range, charge-transfer processes can be observed in batteries**[26]. Electric devices, which are the focus here, are excited by an external stimulus, which allows the introduction of gating of the AC, enabling a more controlled experiment. In particular, this method applies a phase-synchronized portion of the AC waveform during the evolution period of the pulse sequence (Figure 1). In the pulse sequence presented here, a spin echo imaging sequence is used with the evolution period located between the $\frac{\pi}{2}$ and the π pulses. The AC phase is stepped systematically in subsequent measurements, thus producing an indirect time dimension. A Fourier transform along this dimension produces a spectrum containing information about the response of the device to the applied AC voltages. The additional imaging dimensions deliver the spatial localization of these measurements. Results acquired with a range of AC frequencies for a single loop of a wire, a commercial cell and a prototype cell are presented. The comparison between different cells demonstrates that characteristic changes can be observed in such measurements.

Pulse sequence design and theoretical background

This work employs a spin-echo imaging sequence (Multi-Slice-Multi-Echo, Bruker ParaVision) with modifications to allow for encoding of oscillating magnetic fields. The most significant modification is the insertion of a gate pulse in the interval between the $\frac{\pi}{2}$ and π degree pulses which controls the AC current (Figure 1c). Different portions of the AC wave are sampled in this interval in a systematic way, which is defined by its starting point t_n relative to a well-defined point in the AC wave. Experimentally, this is achieved by employing a synchronized square wave triggering the spectrometer to guarantee a start at the same position in the waveform in consecutive cycles. The total encoding time τ is held fixed, but the starting time is shifted by a multiple of the sampling delay $\Delta\tau_{AC}$ in separate experiments (Figure 1a,c). This means that the first segment will be sampled with the interval $[0, \tau]$, while the next segment is sampled with $[\Delta\tau_{AC}, \Delta\tau_{AC} + \tau]$, etc; 16 or 32 of such measurements are typically taken to provide sufficient discrimination between different AC frequencies. The sequence of these experiments allows recording the indirect time dimension along which the AC frequency is encoded.

The AC signal is encoded in the magnetization evolution as follows. After

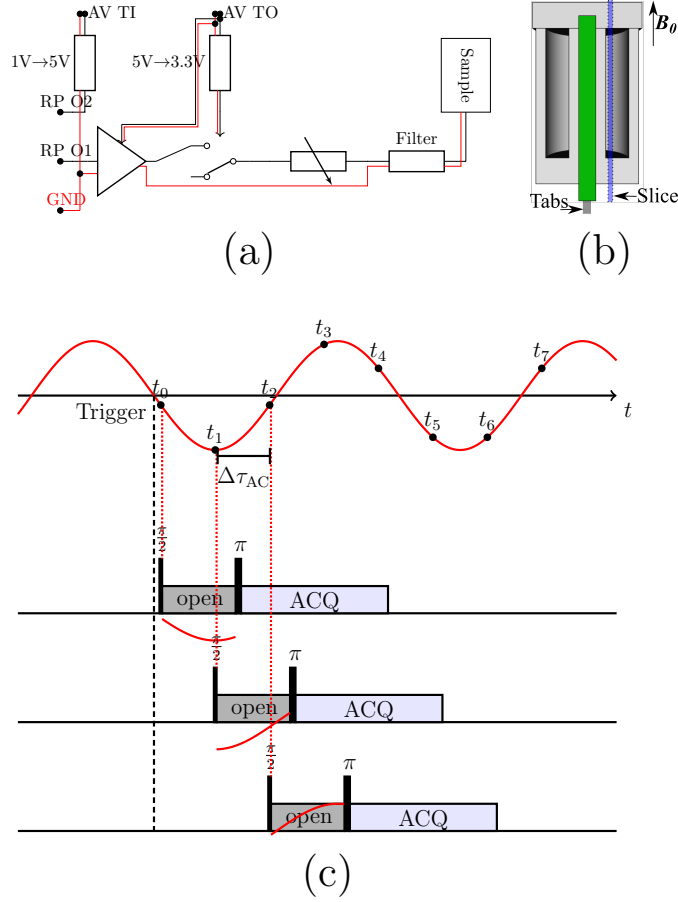


Figure 1: (a) Setup with all electronic components. See Setup and Experimental section for details. (b) 3D-printed holder for the inside-out MRI approach. The battery or coil (green) is located in the middle of the cylindrical holder with two doped water compartments located on each side of the cell/coil. The leads are soldered to the tabs/coil from the bottom of the cell and then guided down the magnet. **The imaging slice is marked by the blue line.** (c) Pulse sequence. A standard spin echo imaging sequence (Multi-slice-multi-echo) was employed. The sequence was modified to allow for sampling through different delay times after the trigger as well as switching the current on and off after/prior to the pulses. In each encoding segment between the pulses, a different phase is encoded, depending on the starting point t_n (depicted exemplary for point 3), which depends on $\Delta\tau_{AC}$ with $t_n = t_0 + n \cdot \Delta\tau_{AC}$.

the excitation pulse to rotate the magnetization into the transverse plane, the magnetization evolves under the influence of the local magnetic field (Figure 1c). The field at a given position $\mathbf{r}$ in space is composed of a static part and a superimposed oscillating field $B_{z,\text{osc}}$. In a frame rotating at the Larmor frequency of the static field ($\omega_0/2\pi$) the signal evolution can be described by

$$S(\mathbf{r}, t_n) = M_0(\mathbf{r}) \cdot \exp \left\{ -i\gamma B_{z,\text{osc}}(\mathbf{r}) \cdot \int_{t_n}^{t_n+\tau} \sin(\omega_{AC} \cdot t') dt' \right\}. \quad (1)$$

$S(\mathbf{r}, t_n)$ is the signal encoded by an oscillating field in the interval $[t_n, t_n+\tau]$ at a position $\mathbf{r}$. M_0 is the bulk magnetization before applying the oscillating field, γ is the gyromagnetic ratio and $B_{z,\text{osc}}(\mathbf{r})$ and ω_{AC} are respectively the magnitude and frequency of the oscillating magnetic field. The time t_n indicates the starting point of the AC wave at the beginning of the evolution period and increments with $\Delta\tau_{AC}$ at each step (as illustrated by the numbered points (0, 1, 2, 3,...) in Figure 1). One can therefore write $t_n = t_0 + n \cdot \Delta\tau_{AC}$, with t_0 being the delay between trigger and AC gating (approximately 6 ms in the experiments done here, the exact delay is not important as long as it is constant).

To simplify discussions, the location-dependence in Equation (1) and associated symbols will be omitted, with the understanding that location-resolved signals will be provided via the imaging portion of the pulse sequence.

The integral can be solved and replaced by an infinite sum of cosine terms via the Jacobi-Anger expansion,

$$S(t_n) = \left(J_0 \left(\frac{\gamma B_{z,\text{osc}}}{\omega_{AC}} \right) + \sum_{k=1}^{\infty} 2i^k J_k \left(\frac{\gamma B_{z,\text{osc}}}{\omega_{AC}} \right) \cos(k \cdot \omega_{AC} \cdot t_n + k \cdot \omega_{AC} \cdot \tau) \right) \cdot \left(J_0 \left(-\frac{\gamma B_{z,\text{osc}}}{\omega_{AC}} \right) + \sum_{l=1}^{\infty} 2i^l J_l \left(-\frac{\gamma B_{z,\text{osc}}}{\omega_{AC}} \right) \cos(l \cdot \omega_{AC} \cdot t_n) \right). \quad (2)$$

As can be seen, a series of oscillating terms at multiples of the base frequency arise. Further details of the derivation are provided in the Appendix. While the series expansion provides conceptual insights, a numerical calculation of Eq. (1) and its Fourier transform was found more practical, and was performed using MatLab (MathWorks, Natick, MA). These simulations obtained for a single frequency and fixed τ show the expected behaviour of peaks

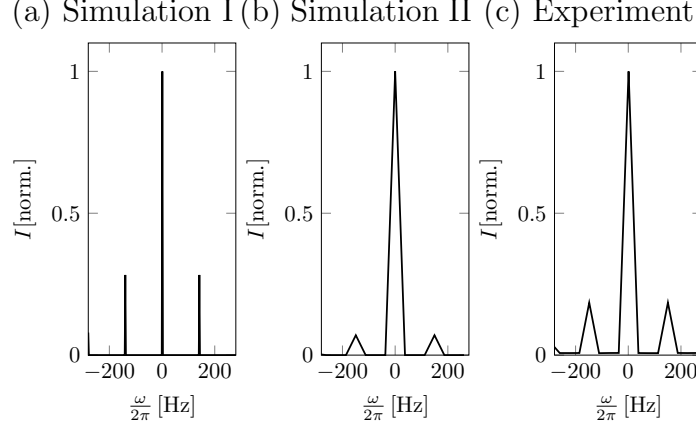


Figure 2: (a): Simulation of the io-TRACING spectrum at 140 Hz AC frequency and 3.21 ms τ with 1024 points of AC sampling and a $\Delta\tau_{AC}$ of 1.786 ms. (b) The same simulation with 16 points (c) Result from an experiment with 16 points of AC sampling. The summed absolute values for each frequency over all pixels in a 2D slice are shown for an experiment with $\omega_{AC}/2\pi=140$ Hz AC frequency and 54 mA AC current.

at multiples of the AC frequency and zero as seen in Figure 2a. Plotting the ratio of the respective frequency peak to the zero-frequency peak versus AC frequency reveals a magnitude sinc-shaped behaviour, i.e. a general decay with AC frequency as well as a substructure leading to zero intensity nodes at multiples of $\frac{1}{\tau}$ (Figure S1). A choice of a small τ changes the position of these nodes, but reduces the encoding strength.

The strong dependence on $\frac{\omega_{AC}}{2\pi}$, with the envelope in Figure S1 decaying with a multiple of $\frac{1}{\frac{\omega_{AC}}{2\pi}}$, also suggests that this method allows the measurement of frequencies of up to approximately 1-2 kHz.

The spectral width for AC-frequency encoding is given by $SW_{AC} = \frac{1}{\Delta\tau_{AC}}$ (Figure 1). As a consequence, this method can be employed to scan for an unknown frequency by employing a small sampling delay and many steps. The acquisition can be tailored to a specific frequency range by adjusting the sampling delay according to this expression. A compromise can often be found between sufficient spectral resolution and experiment duration. This approach is particularly important at low frequencies. For example, for 0.1 Hz, the sampling delay, $\Delta\tau_{AC}$, becomes 2.5 s for a io-TRACING spectral width of 0.4 Hz, and the experiment times for 8, 16 or 32 points in the indirect AC dimension (phase dimension 64 points, 4 scans, TR 125 ms) become roughly

157 6.4 h, 24.2 h and 93.9 h, respectively. The inconvenient lengthening of exper-
 158 iment times at very small frequencies is also known in EIS.

159 The spectral resolution is given by the spectral width for AC-frequency
 160 encoding and the number of encoded segments n_{enc} : $\frac{\text{SW}_{\text{AC}}}{n_{\text{enc}}} = \frac{1}{\Delta\tau_{\text{AC}} \cdot n_{\text{enc}}}$. The
 161 sampling delay should be set to capture all frequencies in the sample, lead-
 162 ing to the conditions $\text{SW}_{\text{AC}} > 2\frac{\omega_{\text{AC}}}{2\pi}$, so $\Delta\tau_{\text{AC}} < \frac{\pi}{\omega_{\text{AC}}}$, and n_{enc} set to distin-
 163 guish between different frequencies, or the frequency and the zero peak, so
 164 $\frac{\text{SW}_{\text{AC}}}{n_{\text{enc}}} < \frac{\Delta\omega_{\text{AC}}}{2\pi} \rightarrow n_{\text{enc}} > \frac{2\pi\text{SW}_{\text{AC}}}{\Delta\omega_{\text{AC}}} \rightarrow n_{\text{enc}} > \frac{2\pi}{\Delta\omega_{\text{AC}}\Delta\tau_{\text{AC}}}$. The encoded signal
 165 (after Fourier transformation along this dimension) is proportional to the
 166 field in the low-field regime for a specific τ (for $\tau=3.21$ ms and $B < 1.5$ μT)
 167 (Figure S3), hence it is a direct measure of the z field strength at a certain fre-
 168 quency. There is a functional relationship between intensity and frequency
 169 (Figure S1), which requires precise calibration with the encoding time τ .
 170 While this parameter can be set, switching times of electronic components
 171 alter the value. Here, a field estimate based on simulations is pursued, which
 172 is inaccurate at frequencies close to $\frac{1}{\tau}$. In principle, experimental calibration
 173 is possible with e.g. a coil fixed in position and immersed in the medium.

174 Setup and experimental

175 The experimental schematic is shown in Figure 1a. The AC waveform
 176 was generated with a Red Pitaya STEMLab 125-14 FPGA controller unit.
 177 The generated signal was amplified with a Stanford Research Systems SR560
 178 Low Noise-Amplifier (Gain 1000). To trigger the spectrometer from the Red
 179 Pitaya control unit, a square wave was generated on the second output of
 180 the unit and then shifted from 1 V to 5 V employing an Arduino UNO. The
 181 amplifier blanking was realized with an output of the spectrometer. In the
 182 case of batteries, a switch (PE42020, Peregrine) was employed to prevent DC
 183 discharge against the output impedance of the amplifier when AC current is
 184 not applied to the battery. This switch is able to reliably disconnect the
 185 battery from the generation at the frequencies employed in this study. The
 186 output is fed to the measurement cell through a variable resistor to fine tune
 187 the current level, and a filter to remove high frequency components in the
 188 RF region.

189 The experiment was set up such that the magnetic fields were mapped
 190 around the battery with a suitable detection medium and MRI sequence
 191 according to the ioMRI strategy [17, 15]. A polylactic acid (PLA) cell holder
 192 (outer diameter 19.75 mm) was 3D-printed consisting of a slot to hold the

193 battery and two semicircular chambers next to that slot (Figure 1b). The
 194 chambers were filled with aqueous 0.1 M CuSO_4 solution. The coil used
 195 in the evaluation experiments consisted of a single loop with a diameter
 196 of 18 mm. Two batteries were employed in this study. (1) A Rochester
 197 Institute of Technology (RIT) cell: Pouch cell Li-ion battery with 250 mAh
 198 capacity. The battery is made of five layers of stacked-cut graphite anode
 199 and NMC cathode electrodes. The cathode is composed of Co (5.97 wt%),
 200 O (19.46 wt%), Ni (14.46 wt%), and Mn (9.19 wt%). (2) Powerstream (PS)
 201 cells: Jelly rolled lithium ions battery with 600 mAh capacity. The graphite
 202 and NMC electrodes are rolled in twelve active layers and packed inside
 203 an aluminum pouch case. Both batteries are made from graphite anode,
 204 aluminum and copper current collectors. The cathode in the PS battery
 205 is made of Co (44.76 wt%), O (33.20 wt%), Ni (4.79 wt%), Mn (2.99 wt%).
 206 Both cells were fully charged prior to the experiment. The dimensions of
 207 the RIT and PS cells are $65.7 \times 42.5 \text{ mm}^2$, and $40 \times 30 \text{ mm}^2$, respectively.
 208 Consequently, the battery tabs of the RIT cell are located further out of the
 209 measurement volume than the PS cells. Since the holder has a 10 mm bottom
 210 to prevent leaking, the lowest 10 mm of the battery close to the tabs are not
 211 represented. Furthermore, on the opposite side, the image is limited by the
 212 excitation volume of the RF coil.

213 A 400 MHz Bruker Avance magnet with a 57 mm gradient system (max.
 214 gradient strength 0.3 T/m) and a 40 mm ^1H coil was employed for the ex-
 215 periments. The battery or coil was rotated with the holder to align parallel
 216 with the B_1 field to minimize artifacts. The main experimental parameters
 217 were set as follows: echo time 10 ms (except stated otherwise); repetition
 218 time 120 ms; number of averages 2; number of repetitions (AC frequency di-
 219 mension, number of different phases sampled) 8 (PS cell $< 0.25 \text{ Hz}$)/16 (PS
 220 cell $> 0.25 \text{ Hz}$, coil)/32 (RIT cell); sampling delay for the AC frequency: $\frac{1}{4 \cdot \frac{\omega_{\text{AC}}}{2\pi}}$
 221 (set with the expected AC frequency, see the section on pulse sequence design
 222 and theoretical background; pulse shapes: sinc3, excitation pulse length 2 ms
 223 (90°); refocusing pulse length 1.267 ms 1.5 ms for coil experiments), field-of-
 224 view (except different orientations) $40 \times 30 \text{ mm}^2$; matrix size $128 \times 64/32$;
 225 slice thickness 2 mm; dwell time 20 μs . With these settings, the refocusing
 226 pulse matches (coil experiment) or exceeds (battery experiment) the excita-
 227 tion pulse bandwidth.

228 The number of samples in the AC dimension is NR, and the increment
 229 of the delay t_n is $\Delta\tau_{\text{AC}}$. For each of these, a full k -space is acquired.

For a two-dimensional image such as those obtained in this study, the data is subject to a three-dimensional Fourier transform, where two dimensions represent image dimensions and one dimension the AC field frequency dimension. Each pixel of the image is then associated with a spectrum of NR points. To identify the dominating frequencies, all pixels are integrated and the three highest peaks are identified. Then, the data point number of e.g. the positive peak is identified, its intensity set in relation to the zero-frequency peak intensity, here referred to as relative intensity $I_{\text{ratio}} = |I_{\frac{\omega_{\text{AC}}}{2\pi}}|/|I_0|$, which then can be plotted as an image.

Evaluation of the technique with a single loop coil

A single-loop coil was used to test the measurement setup and to demonstrate the ability to map the oscillating magnetic fields. According to the presented theoretical considerations, the Fourier transform of the signal features peaks at zero and $\pm n \frac{\omega_{\text{AC}}}{2\pi}$. If the corresponding spectra of all pixels are summed up, a global spectrum at a frequency of $\frac{\omega_{\text{AC}}}{2\pi}$ is obtained, which shows the expected behavior (Figure 2, c). Overtone signal peaks may be observed at lower intensities at multiples of the base frequency.

The spatial distribution of the magnetic field does not change with frequency for the single loop measurements. This is expected as the coil impedance does not change in the frequency range of the applied current (0.1 - 4000 Hz). In the images of a slice parallel to the coil plane, the z -component of the field can be observed with a zero node in the middle of the coil and two regions of high intensity (Figure 3). A plane parallel to the coil at a certain distance is expected to show a z -field, which increases from the center position upwards and then decreases beyond the coil radius (see the simulations in Figure S3). The sign of the field was not determined with this approach due to magnitude processing. The phase of single pixels is frequency dependent, as the Fourier terms in the supporting information reveal, but the dependence is complex. In the future, it may be possible to obtain phase information of the field by employing a phase-sensitive acquisition scheme.

While the pattern of the observed field magnitude remains largely the same as a function of frequency, the overall intensity changes, as expected from numerical simulations of Equation 1 (Figure S1). Furthermore, the encoding strength can be controlled by a change of the echo time, or more precisely the encoding time τ (Figure S4). In the bottom row of Figure 3, the intensity variation with frequency can be observed. To compare field

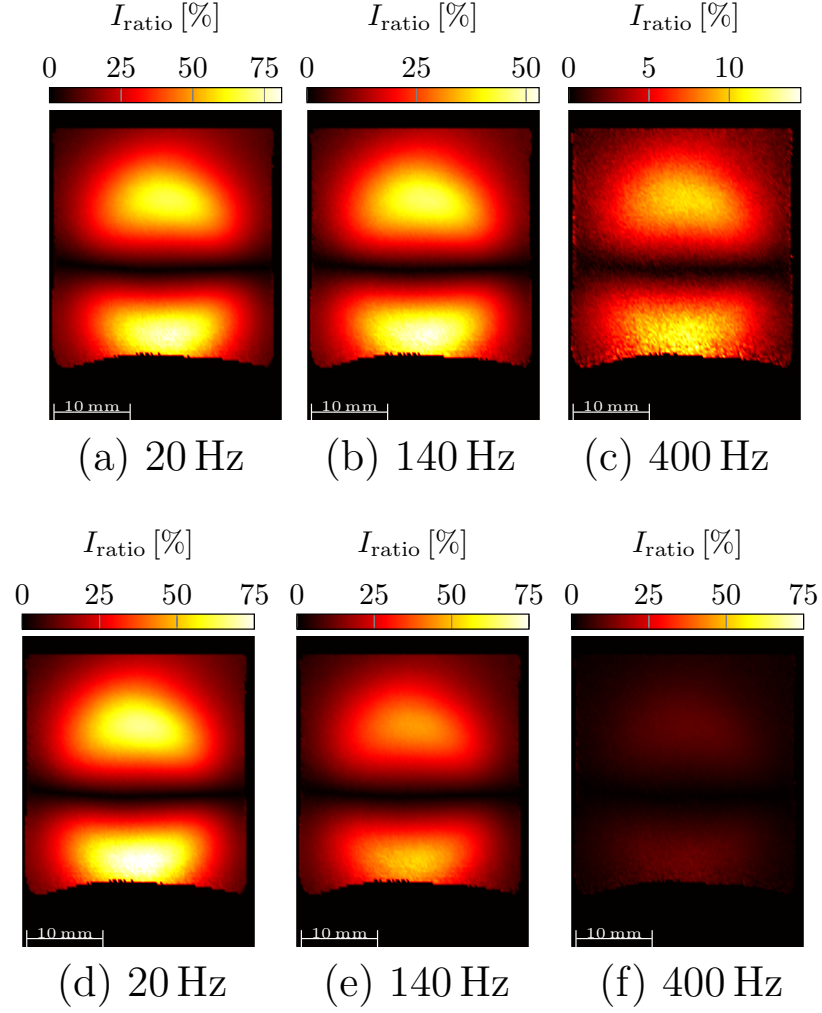


Figure 3: Comparison of the positive frequency part of the spectrum at different AC frequencies with a current of 54 mA. While the absolute intensity changes (bottom row), the relative patterns remain (top row). 16 experiments with different parts of the waveform have been employed for reconstruction resulting in t_n of 0 to 15 $\Delta\tau_{\text{AC}}$.

266 pattern changes, it is desirable to have a common scale. Therefore, the im-
 267 ages have to be multiplied by a factor related to the physical field, which
 268 can be estimated from simulations. The integrated signal ratio between the
 269 first frequency peak at $\omega = \omega_{AC}$ and the $\omega = 0$ peak over the whole image at
 270 these frequencies should therefore follow the simulated response. In Figure 4,
 271 the experimental data agree relatively well with the simulation, although the
 272 encoding time seems to be slightly different due to electronic part switching
 273 times, causing shifts in the pattern. **Although the switching times for the**
 274 **amplifier blanking are given by 5 μ s and 15 μ s, respectively, the best match**
 275 **was found by adding a 60 μ s delay to the simulation.** Furthermore, the in-
 276 tensity of the 20 ms echo time curves do not seem to line up. At such a high
 277 echo, the encoding of field information is strong, which causes inverted ratios
 278 of $\omega = 0$ and $\omega = \omega_{AC}$ peaks and therefore elevated errors. This is enhanced
 279 by the normalization of the intensity in this Figure in order to compare be-
 280 tween simulation and experiment. Smaller encoding/echo times lead to lower
 281 intensity as also shown in Figure S1. Furthermore, as expected, the intensity
 282 is a function of current and scales proportionally (Figure S5).

283 The numerical simulations can also serve as a way to estimate the field.
 284 The ratio of peaks, which has a magnitude sinc-like behaviour with AC fre-
 285 quency as shown in Figures 4 and S1, can be employed to calculate a factor
 286 to transform the data into field estimates. However, due to small differ-
 287 ences in encoding time caused by electronic component switching, the curves
 288 of experiment and simulation do not completely agree with each other and
 289 seem to be shifted (**see above**). Therefore, especially at frequencies close to
 290 the zero-intensity nodes, this method is not very accurate. The peak field
 291 amplitude is estimated to be 1.5 μ T. At half of the current, the intensity is
 292 also approximately halved leading to a field estimate of 0.75 μ T (Figure S5).
 293 A simulation with in-house Matlab code shows the same field pattern and
 294 reproduces the observed field values up to a residual discrepancy at a level
 295 of 0.93 μ T (Figure S3), 7.5 mm slice position). Small differences in sample
 296 positioning **can** lead to strong changes in magnitude **with the current sample**
 297 **arrangement**. Therefore, a small offset of the distance leads to a slightly
 298 larger or smaller value. Evidence for this assumption can be found in Fig-
 299 ure S6 (b), where both sides show different intensities. **While agreement**
 300 **between experiment and simulation is good, more precise positioning will**
 301 **allow better quantification of overall amplitudes and field distributions. In**
 302 **addition, inaccuracies in the field estimate can arise due to division by small**
 303 **values near the nodes in Figure 4.**

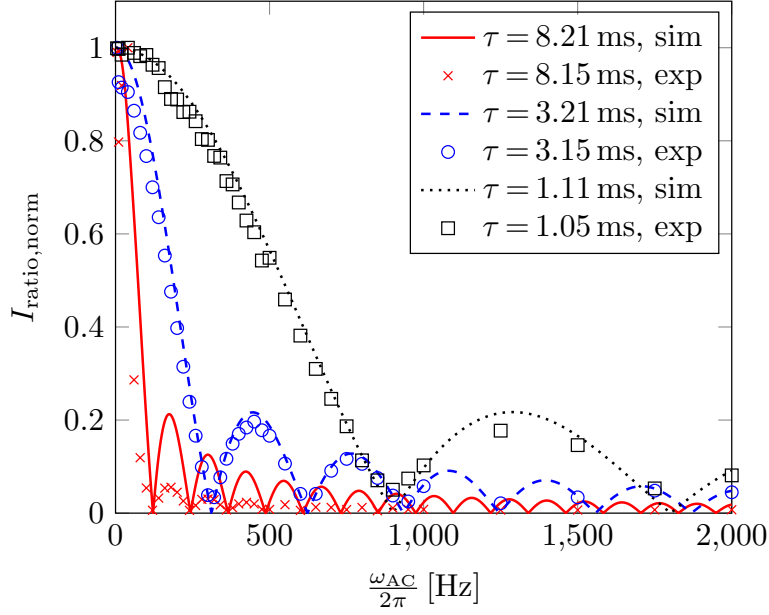


Figure 4: Comparison of experimental (positive frequency, single loop, 54 mA, slice distance from coil approx. 7.5 mm) and simulated data. The experimental data represent an integral of the whole image and all curves have been normalized for comparison purposes. The encoding time for the AC frequency, τ is related to the echo time (TE) via $\tau = \frac{TE}{2} - 1.85$ ms, since the AC field is turned on only during the first echo period, and TE also includes additional pulse durations. 16 experiments with different parts of the waveform have been employed for reconstruction of each point. A delay of 60 μ s has been added to the simulation data to account for imperfections in AC feed current amplifier switching in the experiment. With these adjustments a near-perfect agreement can be obtained, especially at the shorter times. Even the uncorrected curves show very good agreement (see Figure S7)

304 Application of the technique to commercial batteries

305 The battery tabs are located at the top of the images shown in Figure 5.
306 Measurements of a PS jelly-rolled 600 mAh cell show small changes with
307 frequency. A zero node can be observed at all frequencies (Figure 5 (e-h)).
308 One notable observation is that the thickness of the zero node in the images
309 decreases slightly with frequency. Furthermore, a small change of the top
310 pattern can be observed at low frequencies. Another change observed at
311 very low AC frequencies (below 2 Hz) is a broadening of the io-TRACING
312 spectrum peaks to more than one point, leading to minor intensity reductions
313 in the corresponding images. The peak field estimate for this cell is in the
314 range of ~~700~~750 nT.

315 For a RIT cell with 250 mAh capacity, there are significant changes with
316 frequency. While at lower frequencies, the intensity is at the tabs (Figure 5 a),
317 the pattern changes significantly with frequency (Figure 5 b-d). The field is
318 in the range of 300-~~1500~~830 nT.

319 These results are encouraging with respect to demonstrating the ability
320 to detect AC fields originating from cells. The fact that marked differences
321 between the different types of cells are seen shows that battery characteristics
322 do leave their marks on the measurements. The two cell types differ in their
323 designs, capacities, and form factors. Notably one has stacked electrodes, and
324 the other rolled ones. Furthermore, the observation that the pattern changes
325 with frequency for only one of them indicates that the AC measurement holds
326 a lot more information than can presently be extracted.

327 It is recognized, however, that the present work does not constitute proof
328 that the AC measurement provides significant improvements in the diagnostic
329 ability or differentiation between cells. More work with a careful selection
330 of cell types, designs, and properties, would be necessary to delineate the
331 connection between underlying changes in electrochemical behavior and the
332 measured oscillating fields in cells, which exceeds the scope of this article.

333 Future improvements of the method could include acceleration with ad-
334 vanced imaging schemes such as Rapid Acquisition with Relaxation Ena-
335 hancement (RARE)[27], a constant-time implementation to remove the fre-
336 quency dependence on the signal intensity, and a lock-in detection to extract
337 the sign and phase of the field and improve the quantification of the field.
338 The developed methodology could also be applied to devices other than the
339 battery cells considered here, including, for example, fuel cells, and other
340 electrochemical testing chambers in both inside-out and conventional imple-

341 mentations.

342 Conclusions

343 A new technique is presented, allowing measuring oscillating magnetic
 344 fields around battery cells. In contrast to many other techniques requir-
 345 ing synchronization of the AC frequency with the whole pulse sequence, in
 346 the current io-TRACING approach, the AC sampling conditions can be set
 347 independently of the sequence. The technique relies on triggering from a fre-
 348 quency generator as its only input to insure a consistent start point for the
 349 sequence. The method is relatively easy to implement with minor electronic
 350 parts external to the spectrometer. In combination with the inside-out MRI
 351 (ioMRI) approach, complicated systems such as batteries and other devices
 352 can be investigated. In this work, changes between different Li-ion battery
 353 types were observed. The developed technique could be applied to other
 354 devices and processes, and may aid in nondestructive battery characteriza-
 355 tion, by providing information akin to a localized version of electrochemical
 356 impedance spectroscopy.

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 362 funding.

363 Appendix A.

364 *Approximations to the Fourier transform of the signal equation*

365 The integrated signal equation is (with $f_{AC} = \frac{\omega_{AC}}{2\pi}$):

$$S(t_n) = M_0 \cdot \exp \left\{ i \cdot \gamma \frac{B_{z,osc.}}{2\pi f_{AC}} \cdot (\cos(2\pi f_{AC} \cdot (t_n + \tau)) - \cos(2\pi f_{AC} \cdot t_n)) \right\} \quad (A.1)$$

366 This expression can be rewritten with the help of the Jacobi-Anger ex-
 367 pansion

$$\exp(iz \cos \Phi) = J_0(z) + 2 \cdot \sum_{n=1}^{\infty} i^n J_n(z) \cos(n\Phi), \quad (A.2)$$

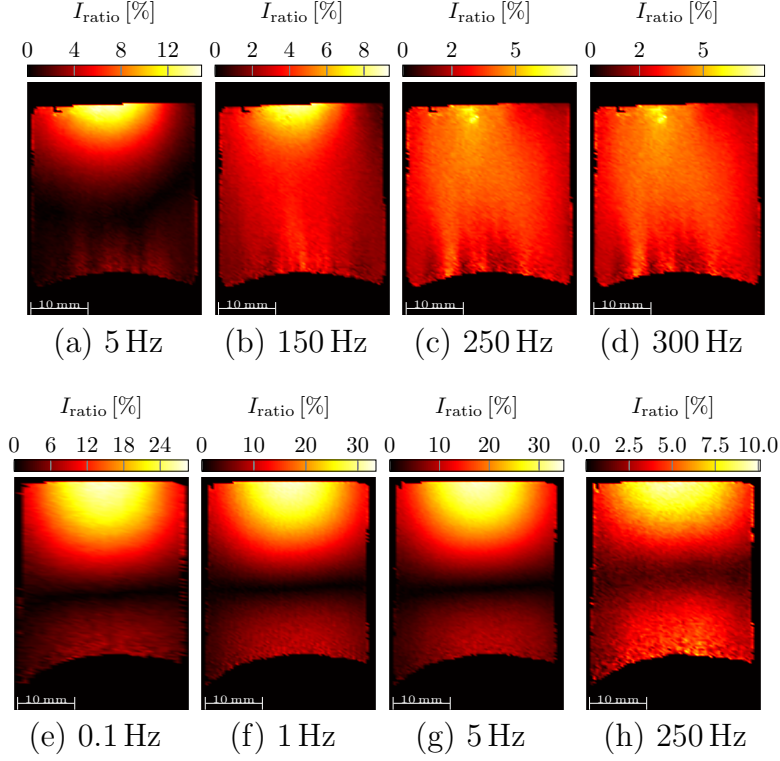


Figure 5: Relative measurements of the oscillating fields generated by 50 mA AC current in various Li-ion batteries at different driving AC frequencies. Slices are parallel to the battery at a distance of approx. 7.5 mm from the center with the battery tabs being located at the top of the image. (a)-(d) are maps from a 250 mAh RIT cell, which has a stacked electrode design. The images in (e)-(h) are from a 600 mAh PS jelly-rolled cell. 16 experiments with different parts of the waveform have been employed for reconstruction of each point of the PS images (t_n from 0 to 15 $\Delta\tau_{\text{AC}}$), while 32 were used for the RIT cell (t_n from 0 to 31 $\Delta\tau_{\text{AC}}$). A different scaled version can be found in Figure S8.

368 which leads to:

$$\begin{aligned}
S(t_n) = & \left(J_0 \left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) + \sum_{k=1}^{\infty} 2i^k J_k \left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) \cos(k \cdot 2\pi f_{\text{AC}} \cdot t_n + k \cdot 2\pi f_{\text{AC}} \cdot \tau) \right) \\
& \cdot \left(J_0 \left(-\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) + \sum_{l=1}^{\infty} 2i^l J_l \left(-\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) \cos(l \cdot 2\pi f_{\text{AC}} \cdot t_n) \right)
\end{aligned} \tag{A.3}$$

369 The Jacobi-Anger expansion consists of a series of Bessel functions. Re-
370 placing the cosine functions with sums of exponentials using the Euler for-
371 mula allows formally writing down the Fourier transformation of this expres-
372 sion. As an example, solutions are provided considering cutoffs of $n \leq 5$ and
373 $m \leq 5$. These expressions are provided for the zero-frequency peak $I(0)$, and
374 the $\pm f_{\text{AC}}$ frequency peak amplitudes $I(\pm f_{\text{AC}})$ are provided below, and a full
375 numerical solution is plotted in Figure S1.

$$\begin{aligned}
I(0) = & \sqrt{2\pi} \{ \\
& e^{-5i\tau 2\pi f_{\text{AC}}} J_0^2 \left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) + (e^{4i\tau 2\pi f_{\text{AC}}} + e^{6i\tau 2\pi f_{\text{AC}}}) J_1^2 \left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) \\
& + e^{3i\tau 2\pi f_{\text{AC}}} J_2^2 \left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) + e^{7i\tau 2\pi f_{\text{AC}}} J_2^2 \left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) \\
& + e^{2i\tau 2\pi f_{\text{AC}}} J_3^2 \left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) + e^{8i\tau 2\pi f_{\text{AC}}} J_3^2 \left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) \\
& + e^{i\tau 2\pi f_{\text{AC}}} J_4^2 \left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) + e^{9i\tau 2\pi f_{\text{AC}}} J_4^2 \left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) \\
& + e^{10i\tau 2\pi f_{\text{AC}}} J_5^2 \left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) + J_5^2 \left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}} \right) \\
& \} \tag{A.4}
\end{aligned}$$

$$\begin{aligned}
I(f_{\text{AC}}) = i\sqrt{2\pi}e^{-4i\tau 2\pi f_{\text{AC}}} \{ \\
& -1 + e^{i\tau 2\pi f_{\text{AC}}} J_0\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_1\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) \\
& - e^{3i\tau 2\pi f_{\text{AC}}} (-1 + e^{3i\tau 2\pi f_{\text{AC}}}) J_2\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_1\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) \\
& + e^{2i\tau 2\pi f_{\text{AC}}} J_2\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_3\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) - e^{7i\tau 2\pi f_{\text{AC}}} J_2\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_3\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) \\
& + e^{i\tau 2\pi f_{\text{AC}}} J_3\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_4\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) - e^{8i\tau 2\pi f_{\text{AC}}} J_3\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_4\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) \\
& - e^{9i\tau 2\pi f_{\text{AC}}} J_4\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_5\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) + J_4\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_5\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) \\
& \} \quad (\text{A.5})
\end{aligned}$$

$$\begin{aligned}
I(-f_{\text{AC}}) = i\sqrt{2\pi}e^{-5i\tau 2\pi f_{\text{AC}}} \{ \\
& - e^{4i\tau 2\pi f_{\text{AC}}} (-1 + e^{i\tau 2\pi f_{\text{AC}}}) J_0\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_1\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) \\
& - e^{3i\tau 2\pi f_{\text{AC}}} (-1 + e^{3i\tau 2\pi f_{\text{AC}}}) J_1\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) \\
& + e^{2i\tau 2\pi f_{\text{AC}}} J_2\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_3\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) - e^{7i\tau 2\pi f_{\text{AC}}} J_2\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_3\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) \\
& + e^{i\tau 2\pi f_{\text{AC}}} J_3\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_4\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) - e^{8i\tau 2\pi f_{\text{AC}}} J_3\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_4\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) \\
& - e^{9i\tau 2\pi f_{\text{AC}}} J_4\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_5\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) + J_4\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) J_5\left(\frac{\gamma B_{z,\text{osc.}}}{2\pi f_{\text{AC}}}\right) \\
& \} \quad (\text{A.6})
\end{aligned}$$

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