Point-by-point response to reviewers and editor for the manuscript:

Innovative L-band electron paramagnetic resonance investigation of solid-state pouch cell batteries, by CE. Dutoit, R. Soleti, JM. Doux, V. Pelé, V. Boireau, C. Jordy, S. Pondaven and H. Vezin.

Reviewer 1

The authors thank the reviewer 1 for having accepted to published this paper in Magnetic Resonance after minor corrections.

Comment #1: *The description of the relaxation time measurements and the values measured should be included in the paper. The response to reviewer comment said that one such number was included in the revised text, but the marked-up copy shows that it was crosses out.*

Response: *Thank you for this comment. Initially and from your comment#9 (interactive discussion), we had modified the relaxation time value T_2 in the revised manuscript. Nevertheless, from the comment#8 of the reviewer 2, about the discussion of the Dysonian line shape, we deleted the sentence "Furthermore, this phenomenon depends on the spin coherent time T2, in which electron travels on a specific distance δe, named spin depth. In metallic lithium structures, T2 ≈10^{-8}s* and then when…".

As suggested by the reviewer 1, we included a short discussion about the relaxation time measurements in the revised version on page 4, lines 86-88.

Comment #2: *The response to reviewers said that they did not observed "local heating in the cell". It is not clear how they would have observed "local" heating "in" the cell. However, one would expect that 0.3mT modulation at 100kHz would produce overall eddy current heating of the aluminum housing and of other conductive elements. This needs to be clarified.*

Response: *Thank you for this point. The comment#2 of the reviewer 2 stated: "… Also, does this high power level cause local heating in the cell?". During measurements, we did not detect any significant heating effects. The EPR spectrum of the Li-metal anode is here similar to the one obtained for a Li-metal sample of similar size wrapped inside a Kapton pouch. Therefore, even though 0.3mT of modulation amplitude at 100kHz can produce eddy current heating the aluminum pouch cell, the effect seems to be negligible. Furthermore, taking the pouch cell with our hands, we did not note that the pouch cell was warmer than before EPR measurements.*

Reviewer 2

The authors have addressed most of my comments in an appropriate way. In particular, sufficient experimental details have been provided to allow reproducing the experiments. There are two points from my initial comments that I still disagree with:

The authors thank the reviewer 2 for having accepted to published this paper in Magnetic Resonance after minor corrections.

Comment #1: *Although I have misunderstood the use of the word "dielectric" (I thought the authors would refer to the sample, not the resonator as detailed in their response), I still think the word should*

be replaced, assuming that the loop-gap resonator does not employ a dielectric? The observation that the resonator mode changes is undisputed, my comment refers only to the use of the word "dielectric".

Response: *Thank you for this comment.* As suggested by the reviewer 2, we replaced the word "dielectric" by "resonator" and we modified the sentence: "Indeed placing a large metallic conductor inside a standard X-band microwave cavity can cause serious perturbations of the dielectric making it impossible to tune the cavity "(on page 4, lines 94) by:

Indeed, placing a large metallic conductor inside a standard X-band microwave cavity can cause serious perturbations of the resonator making it impossible to tune the cavity.

Also, we modified the sentence: "As a result, the dielectric is slightly perturbed by inserting the metallic sample (see Figure 1(b))" (on pages 4-5, lines 98-99) by:

As a result, the microwave resonator is undisturbed by inserting the metallic sample (see Figure 1(b))

Comment #2: *I still fail to imagine a 3D geometry that would explain all three 2D images at the same time. Considering for simplicity, but without loss of generality, that the image can be represented by four bars along the edges, as suggested by the xz image. Then the yz image suggests that all four bars must be located within a plane that is slightly tilted. Such a tilted geometry would explain why the xy image is not simply a single line as well. However, the actual xy image, consisting mainly of two bars that are not parallel to each other, would not result in the displayed narrow line in the yz image. This discrepancy should be discussed in the text.*

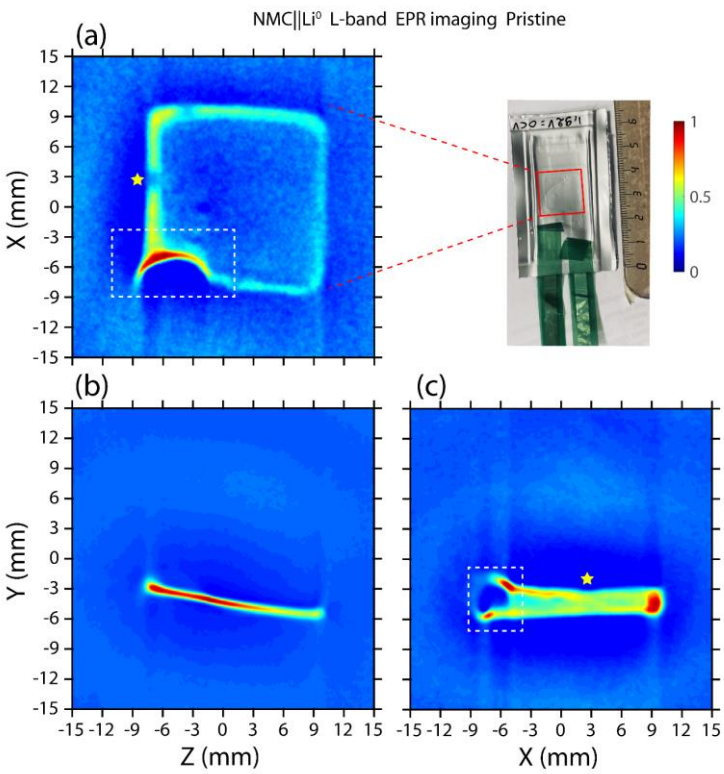
Response: *Thank you for this point. A 3D image reconstruction from three 2D images (not recorded during measurements, but reconstructed from 2D images) can provides a detailed location of metallic lithium aggregates and lithiated graphite complexes but without providing more chemical information. It can be seen as a complementary image.*

All three 2D images were recorded without modifying the physical position of the pouch cell inside the resonator. Therefore, the "irregularities" observed on xz, xy and zy images come from the metallic anode and/or the current collectors. From all three 2D images, it can be seen the pouch cell is tilted in the yz plane and the xy plane. However, the "line" observed in the yz plane, corresponding to the anode electrode, indicates that the pouch cell is aligned in the x direction.

We are in agreement with the reviewer that our xy displays a weak irregularity around X=-6mm and Y=-3mm. The origin of this irregularity comes from the current collector with a semi-circle shape clearly visible in the xz image (X=-6mm and Z between -6mm and -3mm). In the limit of resolution, this current collector is aligned to the yz plane but not in the xz plane (as indicated by the dashed square in the figure below). This point is clarified in the revised version on page 7.

The semi-circle shape is also clearly visible at X=-6 mm and Y=-3 mm in the XY plane but not in the YZ plane indicating that the pouch cell is aligned in the X direction

Furthermore, the metallic anode is not a perfect square but exhibits some surface roughness which explain why we do not observe perfect parallel bars.


NMC||Li⁰ L-band EPR imaging Pristine
(a)
(b)
(c)
X (mm)
Y (mm)
Z (mm)
X (mm)
1
0.5
0