

Supplementary information

Second harmonic electron paramagnetic resonance spectroscopy and imaging reveal metallic lithium depositions in Li-ion batteries

Charles-E. Dutoit^{1,2}, Hania Ahouari^{1,3}, Quentin Denoyelle⁴, Simon Pondaven⁵ and Hervé Vezin^{1,2}

¹Université Lille Nord de France, CNRS UMR8516, LASIRE, 59655 Villeneuve d'Ascq, France

²Centre de Résonance Magnétique Electronique pour les Matériaux et l'Energie, 59655 Villeneuve d'Ascq, France

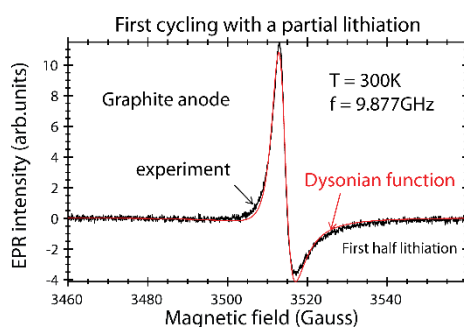
³Université de Lille, FR 2638-IMEC-Institut Michel-Eugène Chevreul, 59655 Villeneuve d'Ascq, France

⁴SAFT, Corporate Research, 111 Boulevard Alfred Dancy, 33074 Bordeaux, France

⁵TotalEnergies, chemin du canal, 69360 Solaize, France

email: charles.dutoit@univ-lille.fr

Supplementary Figure S1: 1st harmonic X-band EPR spectrum of graphite anode recorded after the first half lithiation (electrode potential of 86mV). The red line represents the simulation using a single dysonian function and shows that the experimental spectrum is correctly reproduced suggesting only one contribution.



Supplementary Figure S2: **a** Voltage and current of the symmetric Li-metal electrochemical cell as a function of the cycling time. **b** Set of X-band EPR spectra of the cell recorded before cycling (i) and after the short circuit (ii). Both EPR signals (ii) were obtained using the 1st and the 2nd harmonic detection scheme respectively.

