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In Situ Tuning of Magnetization via Topotactic Lithium Insertion in Ordered Mesoporous Lithium Ferrite Thin Films

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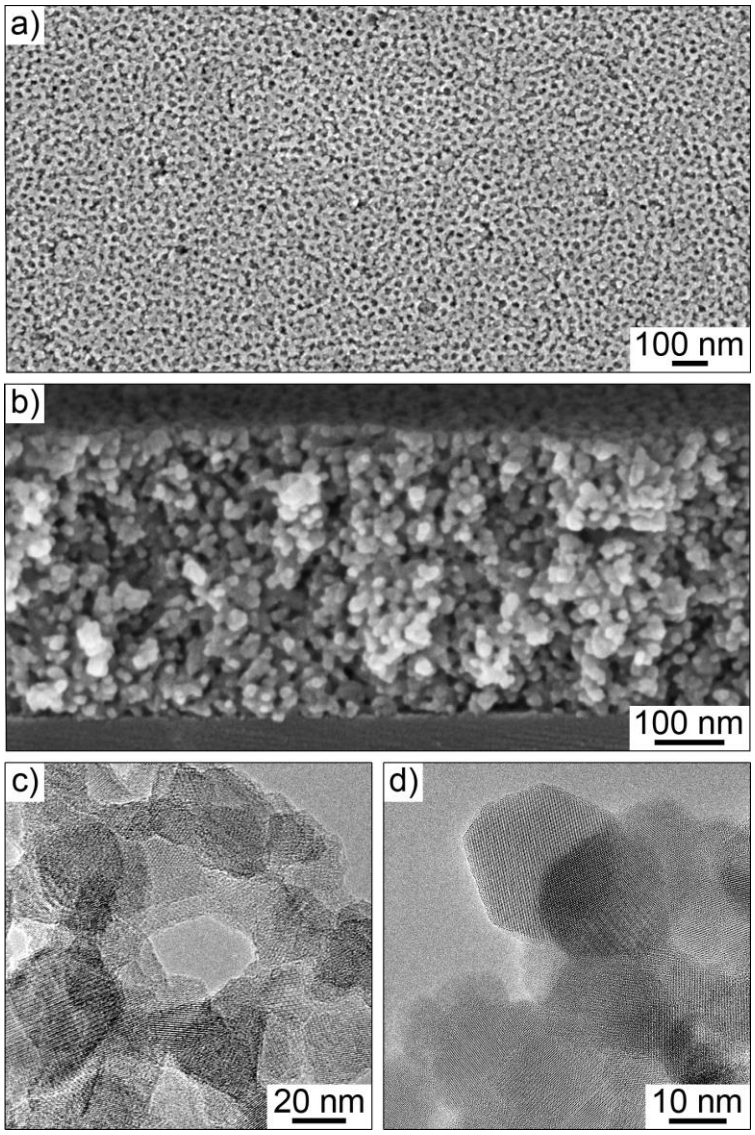

Figure S1. Electron microscopy of mesoporous LFO thin films heated to 630 °C. (a) Top view and (b) cross-sectional SEM images showing that the sol-gel derived material is free of cracks on the micrometer level and the cubic pore structure persists throughout the bulk. HRTEM images collected (c) before cycling and (d) after 20 cycles in a lithiated state. The material remains crystalline and the microstructure does not change when cycled in the potential range from 3.2 to 1.4 V with respect to Li^+/Li.

Table S1. Peak frequencies (cm^{-1}) and Raman band assignment.[1]

	α-$LiFe_5O_8$ single crystal at 300 K	β-$LiFe_5O_8$ single crystal at 300 K	α-$LiFe_5O_8$ porous film at 298 K
1	127 A_1 (1)		125
2	132 E (1)		-
3	176 E (2)		(173)
4	190 F_2 (1)		(188)
5	203 F_2 (2)	201 F_{2g} (1)	199
6	221 F_2 (3)		-
7	236 E (3)		236
8	260 F_2 (4)		261
9	262 E (4)		
10	270 A_1 (2)		-
11	286 F_2 (5)		-
12	300 E (5)		298
13	318 E (6)		-
14	334 F_2 (6)		-
15	359 E (7)	360 E_g (1)	355
16	382 E (8)		378
17	401 A_1 (3)		396
18	410 F_2 (7)		-
19	441 F_2 (8)		436
19	447 A_1 (4)		445
20	471 F_2 (9)		(466)
21	494 F_2 (10)	494 F_{2g} (2)	489
22	519 E (9)		*

23	555 F_2 (11)		(550)
24	611 F_2 (12)	609 F_{2g} (3)	606
25	616 A_1 (5)		-
25	684 F_2 (13)		680
26	714 A_1 (6)	712 A_{1g} (1)	710

* superimposed, () weak

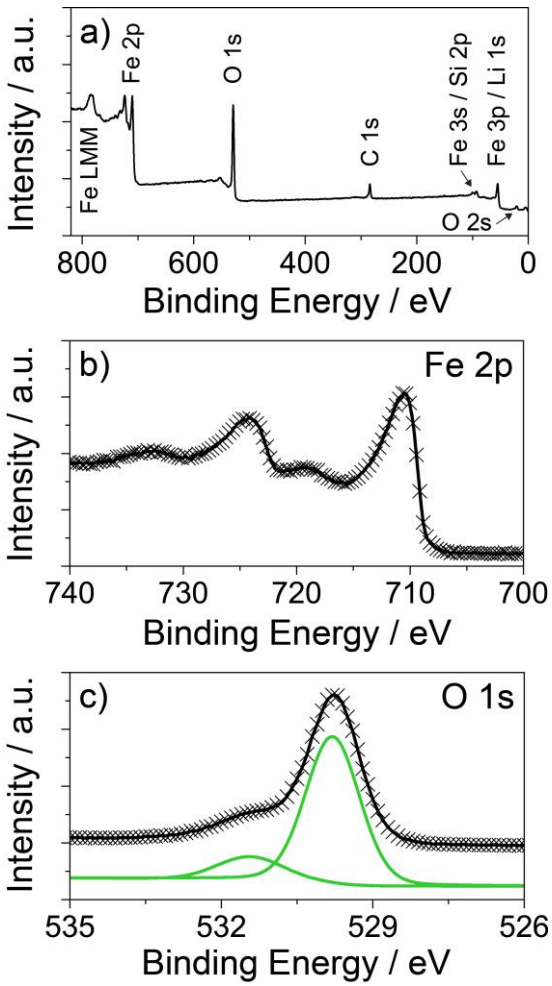

Figure S2. XPS analysis of mesoporous LFO thin films on Si(001) substrate heated to 630 °C. (a) Survey spectrum showing (apart from weak C 1s and Si 2p peaks) only Fe, Li and O levels. (b) Fe 2p spectrum. The presence of satellites around 8 eV higher in binding energy than the main peaks at (710.50 ± 0.1) eV and (724.10 ± 0.1) eV for the $2p_{3/2}$ and $2p_{1/2}$ levels, respectively, is characteristic of the Fe(III) oxidation state. (c) O 1s spectrum indicating two different bonding states. The main peak at (529.80 ± 0.1) eV corresponds to oxygen in LFO and the shoulder peak at (531.4 ± 0.1) eV can be attributed to hydroxyl oxygen.

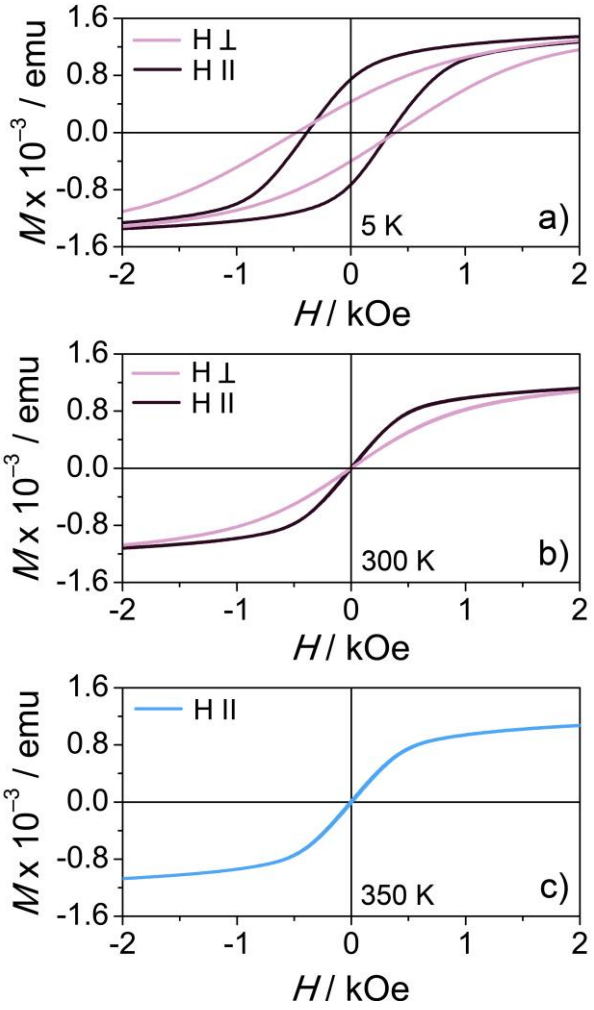

Figure S3. Magnetic properties of mesoporous LFO thin films. Hysteresis curves measured at (a) 5 K and (b) 300 K with the magnetic field oriented parallel as well as perpendicular to the substrate plane. (c) In-plane hysteresis curve indicating superparamagnetic behavior at 350 K.

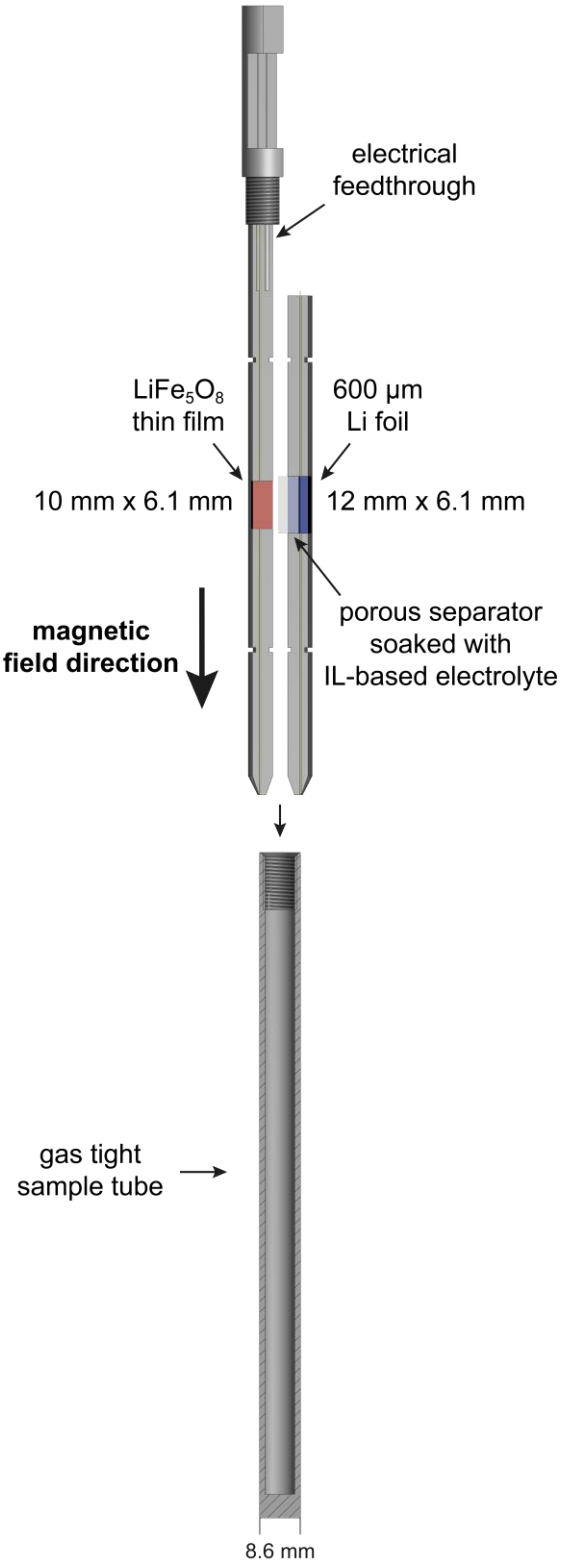

Figure S4. 2D CAD drawing of the home-built cell for in situ magnetization tuning. The wiring was done using 0.1 mm diameter Au wires. The Li electrode was chosen to be slightly larger than the $LiFe_5O_8$ electrode in order to ensure homogeneous current distribution. The cell assembling was performed inside an argon-filled glovebox. The magnetic field direction was set to be parallel to the film surface.

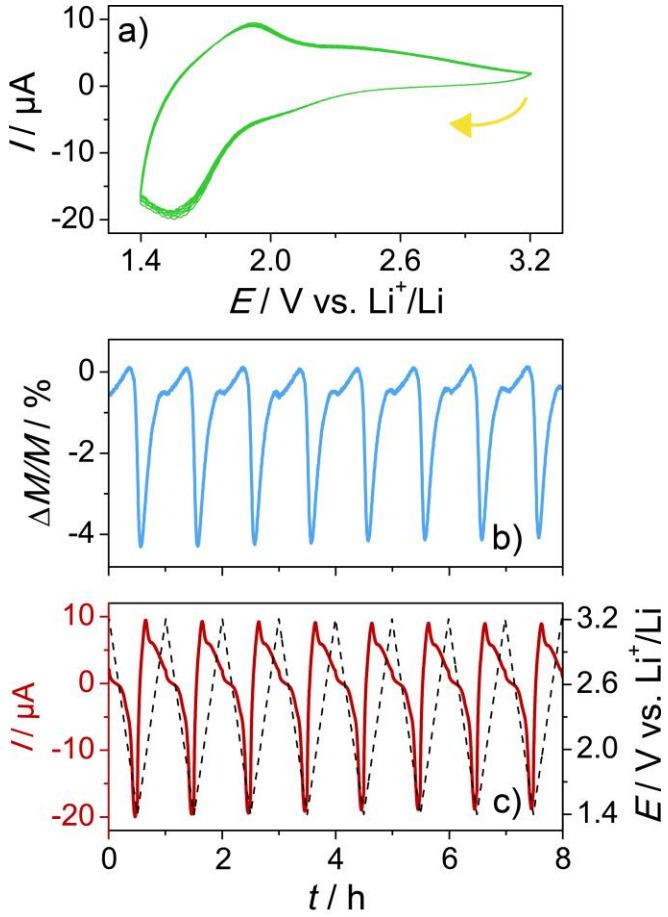

Figure S5. (a) Voltammetry sweeps (8 cycles) at 1 mV/s. (b) Relative change in magnetization measured at room temperature and in an applied field of 100 Oe. (c) Potential and current versus the time.

1. M. N. Iliev, V. G. Ivanov, N. D. Todorov, V. Marinova, M. V Abrashev, R. Petrova, Y. Q. Wang, and A. P. Litvinchuk, *Phys. Rev. B*, 2011, **83**, 174111.