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Electrical, Electronic and Magnetic Properties Correlation Via Oxygen Vacancy Filling and Scaling-Law Analysis in $LiFe_5O_8$ Thin Films Prepared by Pulsed Laser Deposition

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Experimental section

Thickness of the $LiFe_5O_8$ were measured using HRSEM cross-section image with the help of imageJ software. Conductivity of all the thin films were measured using two probe setup with the help of impedance analyzer. Electrode deposition for the impedance measurements were carried out by e-beam evaporation technique. Electrode length, width and the distance between the electrodes is 2 mm, 2mm and 2 mm respectively. We applied the polarization voltage of 0.5 V and frequency of 20 MHz to 1 Hz.

Result and discussion

Figure S1 (a) to (d) shows the XPS survey spectra of all $LiFe_5O_8$ thin films in the thickness range from 60 to 118 nm which confirms that Li, Fe and O present all thin films. Figure S3 (e) to (h) shows the Li 1s spectra of all thin films which indicates that Li present in 1+ state. Figure S2 (a) to (d) shows the Fe 2p core level spectra of $LiFe_5O_8$ thin films in the thickness range from 60 to 118 nm. From the spectra we can confirm that Fe^{3+} and Fe^{2+} ions co-existence in all the thin films. Figure S4 (e) to (h) shows the O 1s core level spectra of all the thin films which indicates that oxygen vacancy decreases with increasing thickness. From Li core level XPS spectra we can observe that the area of Li^+ decreases and Fe 3p area increases which is due to the increase in the Fe content with increasing thickness. XPS studies reveals that oxygen vacancy decreases with and lattice oxygen increases with increasing thickness from 60 nm to 135 nm. These results directly correlated with the saturation magnetization, bandgap and valance band spectra values described in the manuscript. Figure S3 shows the bandgap value of $LiFe_5O_8$ bulk material and found to be 1.79 eV.

Figure S1

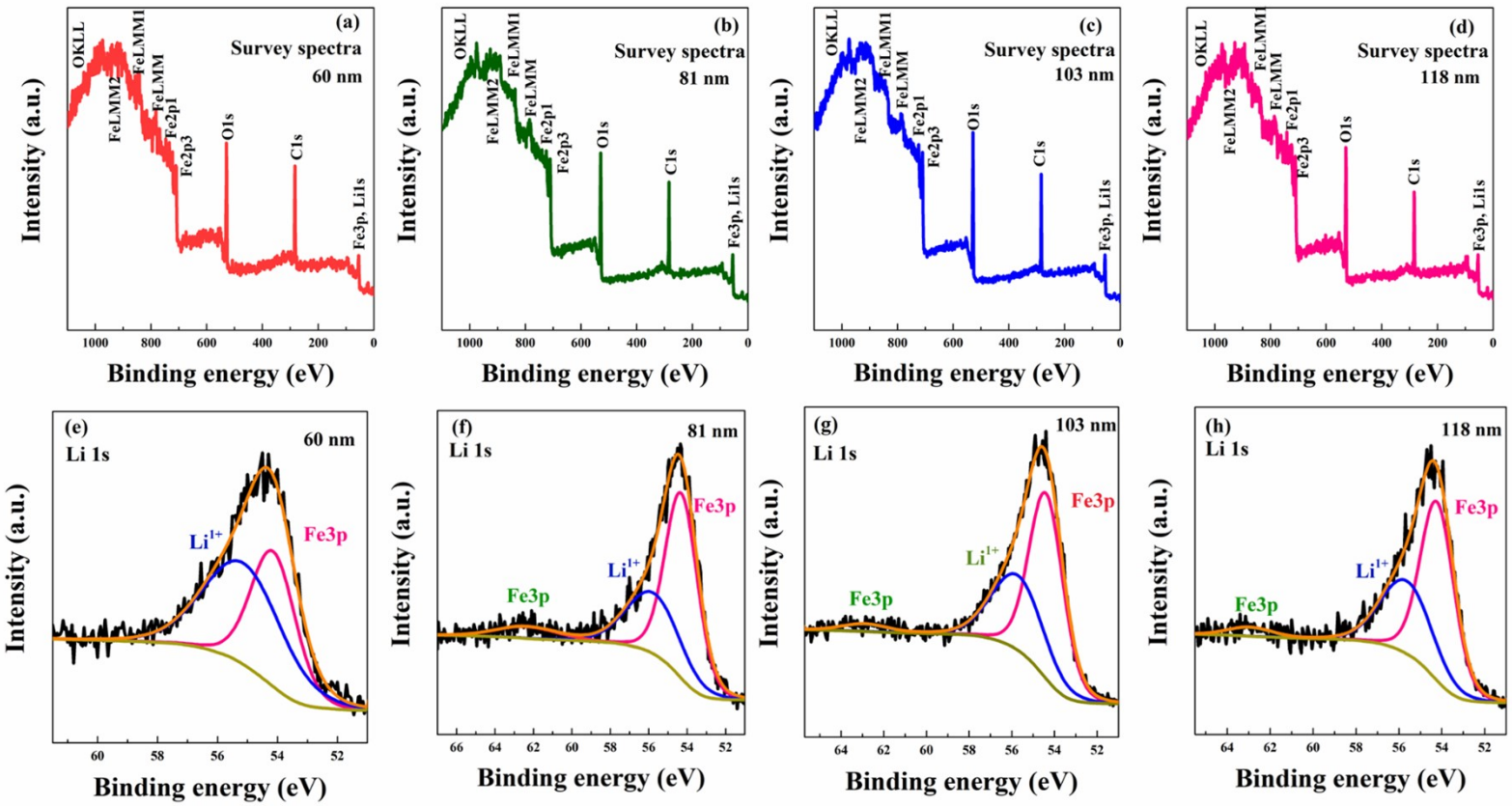

(a)
Survey spectra
60 nm
(b)
Survey spectra
81 nm
(c)
Survey spectra
103 nm
(d)
Survey spectra
118 nm
OKLL
FeLMM2
FeLMM1
FeLMM
Fe2p1
Fe2p3
O1s
C1s
Fe3p, Li1s
Intensity (a.u.)
Binding energy (eV)
(e)
Li 1s
60 nm
(f)
Li 1s
81 nm
(g)
Li 1s
103 nm
(h)
Li 1s
118 nm
Li1+
Fe3p

Figure S2

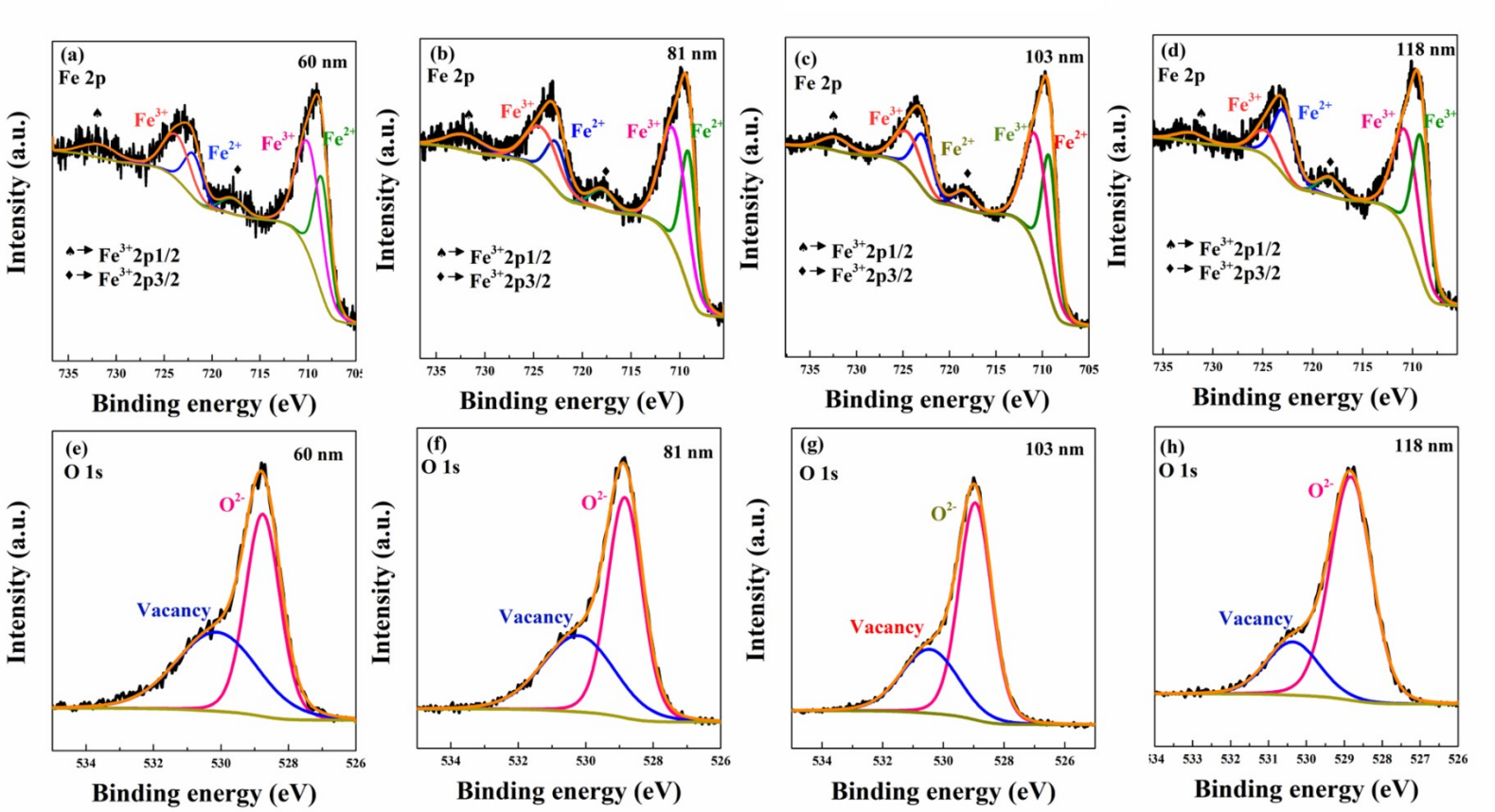
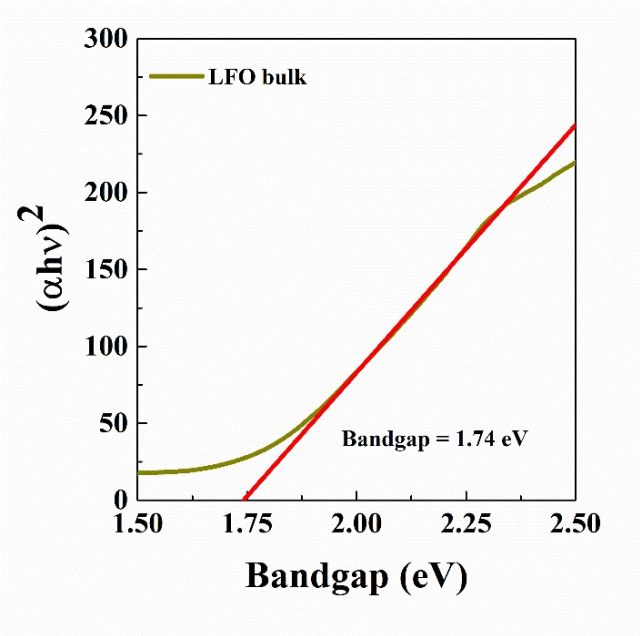

(a)
Fe 2p
60 nm
(b)
Fe 2p
81 nm
(c)
Fe 2p
103 nm
(d)
Fe 2p
118 nm
Fe3+
Fe2+
Fe3+2p1/2
Fe3+2p3/2
Intensity (a.u.)
Binding energy (eV)
(e)
O 1s
60 nm
(f)
O 1s
81 nm
(g)
O 1s
103 nm
(h)
O 1s
118 nm
O2-
Vacancy

Figure S3

Bandgap measured for the bulk $LiFe_5O_8$ material



With increasing the thickness of $LiFe5O_8$ beyond 135 nm i.e to 156 nm, we have observed that the saturation magnetization value increases and the band gap value decreases towards the bulk material value. XRD, SEM, XPS, Magnetization, band gap and the VB plots of 156 nm thin film is shown in Figure S4, S5 and S6 respectively. Room temperature saturation magnetization value is found to be 268 emu/cc and 243 emu/cc for in-plane and out of plane respectively, which clearly supports the increasing trend of saturation magnetization. Bandgap and valance band maxima of the prepared 156 nm thin film is found to be around 2.04 eV and 0.14 eV, which clearly supports the decreasing trend of bandgap of the material towards the bulk material value. The magnetic and band edge parameters are tabulated in Table ST1

Figure S4

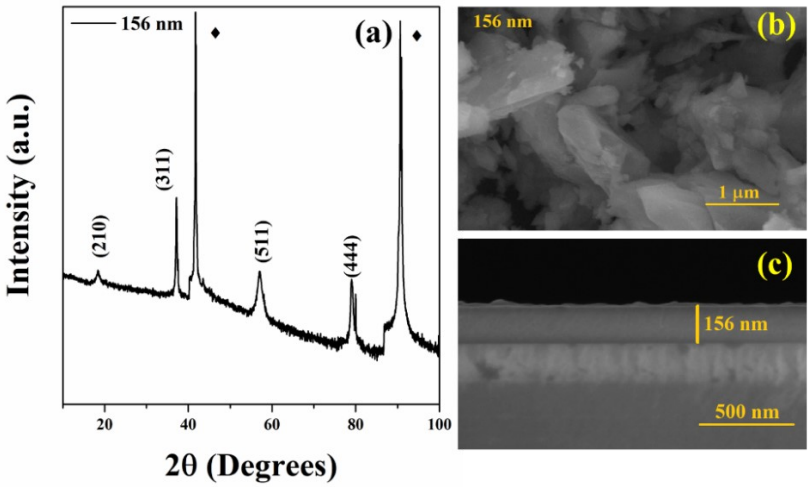


Figure S5

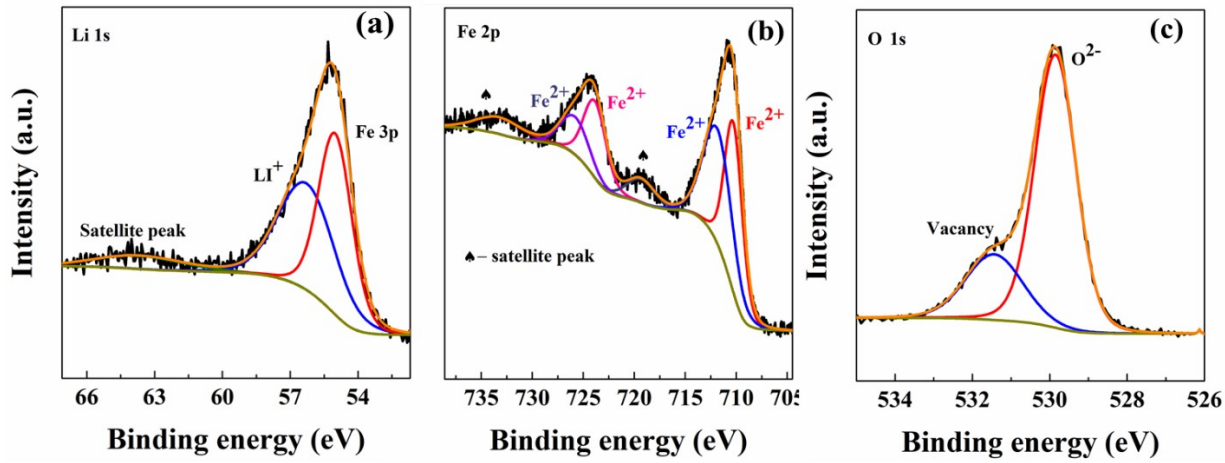


Figure S6

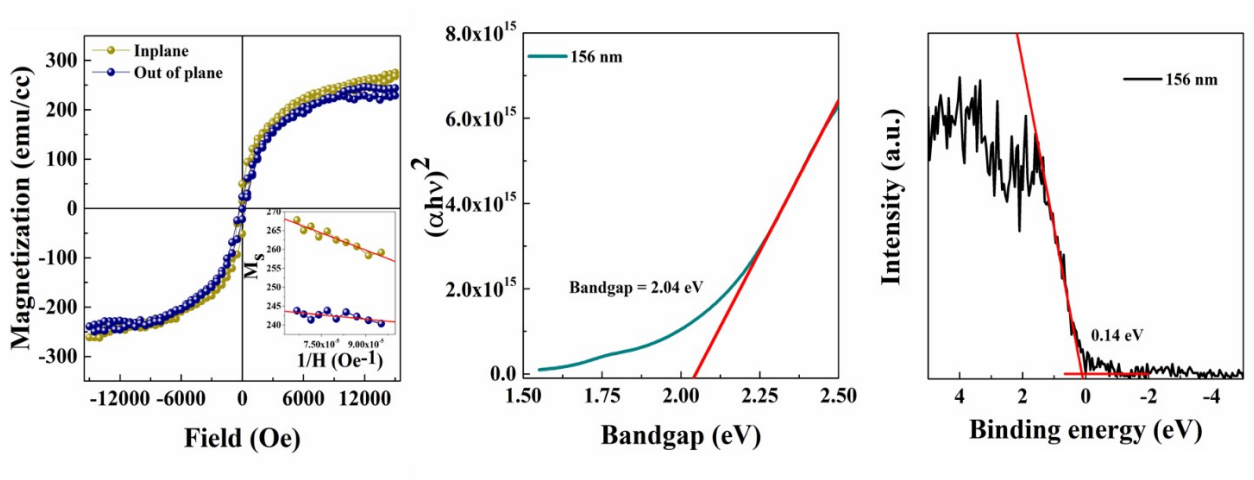


Table ST1

Sample	In plane M_S (emu/cc)	Bandgap (eV)	VBS (eV)
60 nm	152	2.64	0.53
81 nm	157	2.62	0.51
103 nm	165	2.43	0.39
118 nm	184	2.41	0.35
135 nm	225	2.24	0.18
156 nm	268	2.04	0.14