Supporting Information-
A lattice dynamical approach for finding the lithium superionic conductor Li_3ErI_6

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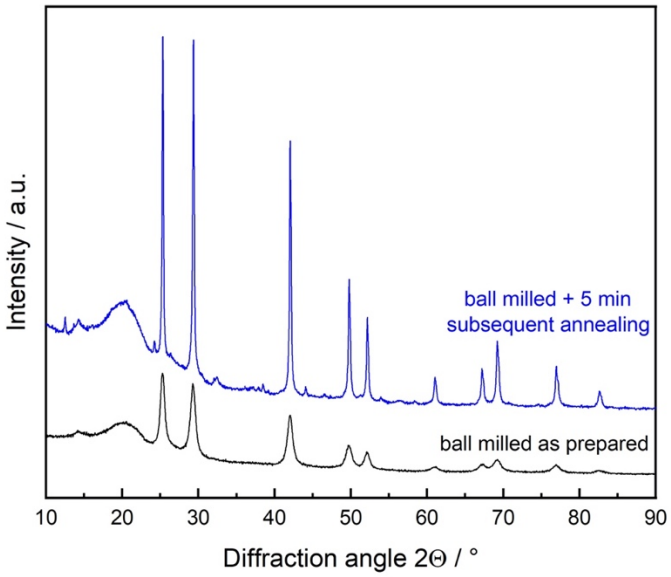

Figure S1: Comparison between the Li_3ErI_6 sample right after ball milling and after an additional 5 min annealing step. The reduction of the background and sharper reflections support the expected increase in crystallinity. This crystallized sample was used for the synchrotron diffraction data collection.

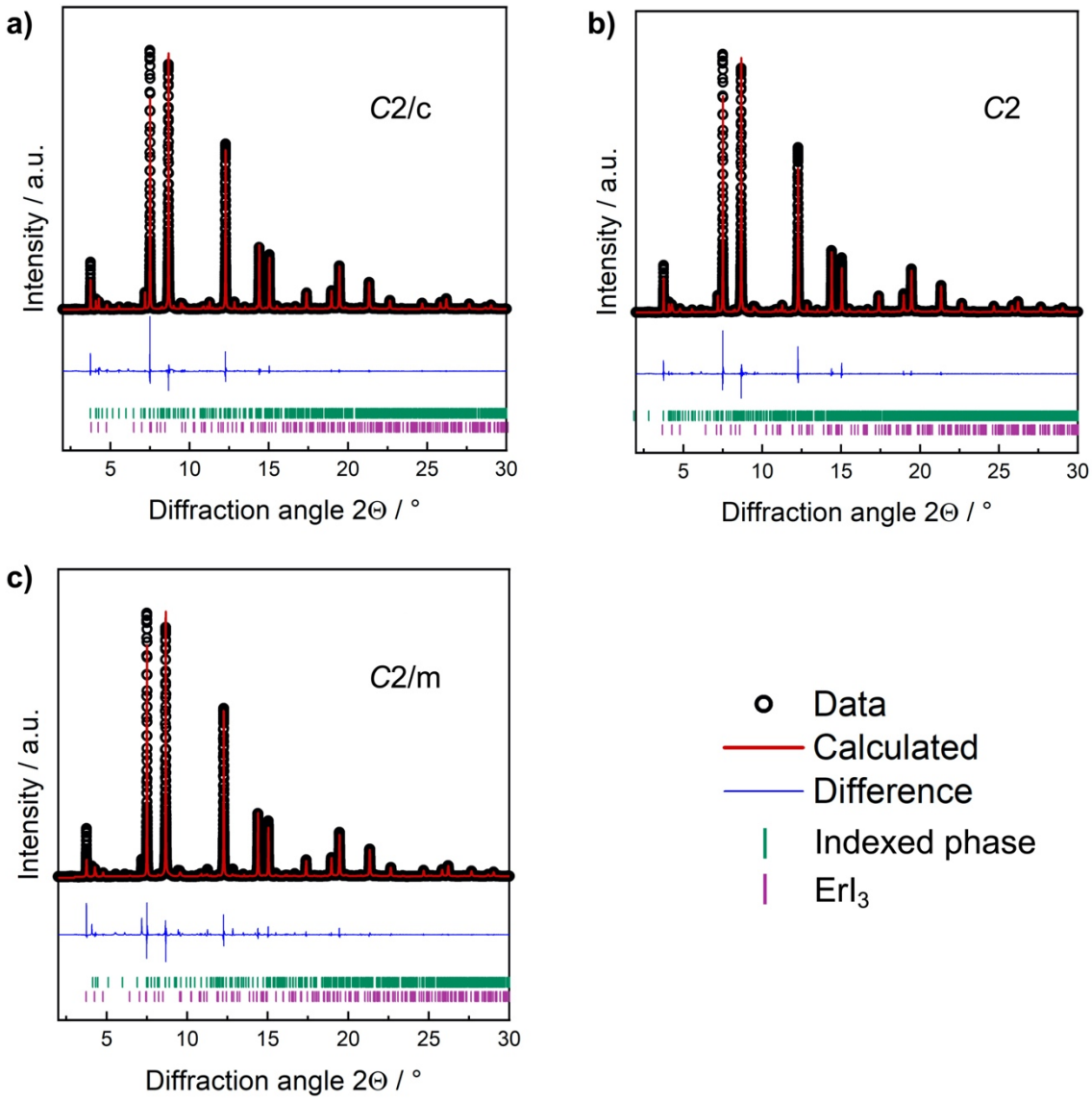

Figure S2: Comparison of Pawley-fits against the collected synchrotron X-ray diffraction data using the different possible space groups. Here, only the space groups C2/c and C2 match all the observed reflection positions, further suggesting one of these being the correct space groups.

Table S1: Used constraints for the synchrotron X-ray diffraction data.

Atom	Wyckoff position	Atomic coordinates			*Occ.*	B_{iso} / Å^2
		x	*y*	*z*		
Er1	4*d*	0.25	0.25	0.5	*1-2·occ1*	*Var1*
Er2	8*f*	0.25	pos1	1	*occ1*	*Var1*
I1	8*f*	pos2	pos5	pos8	1	*Var2*
I2	8*f*	pos3	pos6	pos9	1	*Var2*
I3	8*f*	pos4	pos7	pos10	1	*Var2*

Table S2: Rietveld-Refinement results against synchrotron diffraction data, using space group C2/c.

Li_3ErI_6 synchrotron diffraction data

Lattice parameter: a = 7.4215(2) b = 12.8509(2), c = 14.2130(3), β = 99.98(2) °

R_{WP}= 12.1 %, R_{exp}= 3.6 %, Goodness of fit: 3.4

Impurity phase: 4 wt% total of LiI and ErI_3

Atom	Wyckoff position	Atomic coordinates			*Occ.*	B_{iso} / Å^2
		x	*y*	*z*		
Er1	4*d*	0.25	0.25	0.5	0.6919(8)	2.05(3)
Er2	8*f*	0.25	0.4173(4)	1	0.1540(8)	2.05(3)
I1	8*f*	0.1241(5)	0.4169(3)	0.6260(3)	1	1.87(1)
I2	8*f*	0.1172(3)	0.2489(4)	0.1222(1)	1	1.87(1)
I3	8*f*	0.1136(5)	0.0903(3)	0.6207(3)	1	1.87(1)

Table S3: Used constraints for neutron diffraction data.

Atom	Wyckoff position	Atomic coordinates			*Occ.*	B_{iso} / Å^2
		x	*y*	*z*		
Er1	4*d*	0.25	0.25	0.5	*1-2·(1-occ1)*	*Var1*
Li1	4*d*	0.25	0.25	0.5	*1-(1-2·(1-occ1))*	*Var1*
Er2	8*f*	0.25	pos1	1	*1-occ1*	*Var1*
Li2	8*f*	0.25	pos1	1	*occ1*	*Var1*
I1	8*f*	pos2	pos5	pos8	1	*Var2*
I2	8*f*	pos3	pos6	pos9	1	*Var2*
I3	8*f*	pos4	pos7	pos10	1	*Var2*
Li3	4*e*	0	pos1	0.25	*occ2*	*Var3*
Li4	4*e*	0	pos11	0.25	*1-occ2*	*Var3*

Table S4: Rietveld-Refinement results against neutron diffraction data, using space group C2/c.

Li_3ErI_6 neutron diffraction data

Lattice parameter: a = 7.413(4) b = 12.838(6), c = 14.200(7), $\boldsymbol{\beta}$ = 100.01(5) °

R_{WP}= 1.5 %, R_{exp}= 1.1 %, Goodness of fit: 1.4

Impurity phase: 5 wt% total of LiI and ErI_3

Atom	**Wyckoff position**	**Atomic coordinates**			***Occ.***	***Biso* / Å^2**
		x	***y***	***z***		
Er1	4*d*	0.25	0.25	0.5	0.68(2)	3.4(1)
Li1	4*d*	0.25	0.25	0.5	0.32(2)	3.4(1)
Er2	8*f*	0.25	0.4218(8)	1	0.16(2)	3.4(1)
Li2	8*f*	0.25	0.4218(8)	1	0.84(2)	3.4(1)
I1	8*f*	0.114(3)	0.419(1)	0.627(1)	1	1.15(2)
I2	8*f*	0.116(2)	0.247(2)	0.123(1)	1	1.15(2)
I3	8*f*	0.109(3)	0.0988(8)	0.617(1)	1	1.15(2)
Li3	4*e*	0	0.4218(8)	0.25	0.2(1)	5(1)
Li4	4*e*	0	0.725(6)	0.25	0.8(1)	5(1)

Table S5: Rietveld-Refinement results against neutron diffraction data without the 4th lithium position, using space group C2/c.

Li_3ErI_6 neutron diffraction data

Lattice parameter: a = 7.411(4) b = 12.843(8), c = 14.201(8), $\boldsymbol{\beta}$ =100.01(7) °

R_{WP}= 1.5 %, R_{exp}= 1.1 %, Goodness of fit: 1.4

Impurity phase: 5 wt% total of LiI and ErI_3

Atom	**Wyckoff position**	**Atomic coordinates**			***Occ.***	***Biso* / Å^2**
		x	***y***	***z***		
Er1	4*d*	0.25	0.25	0.5	0.57(3)	0.3(3)
Li1	4*d*	0.25	0.25	0.5	0.43(3)	0.3(3)
Er2	8*f*	0.25	0.4218(8)	1	0.22(1)	0.3(3)
Li2	8*f*	0.25	0.4218(8)	1	0.78(1)	0.3(3)
I1	8*f*	0.114(3)	0.419(1)	0.627(1)	1	1.24(4)
I2	8*f*	0.116(2)	0.247(2)	0.123(1)	1	1.24(4)
I3	8*f*	0.109(3)	0.0988(8)	0.617(1)	1	1.24(4)
Li3	4*e*	0	0.4218(8)	0.25	1	1.0(7)