Supplementary Information

Neutron diffraction study of the tetragonal – monoclinic phase transition in $NdNbO_4$ and $NdTaO_4$

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Additional Experimental Details

Temperature Ramp Rates:

Sample NdTaO4

RT – 500 ^{o}C 1K/min: 500-1500 ^{o}C 0.5 K/min; 1500-650 ^{o}C 0.5 K/min 650 – 150 ^{o}C 1 K/min

Sample NdNbO4

RT – 500 ^{o}C 1 K/min: 500-1000 ^{o}C 0.5 K/min; 1000-750 ^{o}C 1 K/min 750 – 550 ^{o}C 0.5 K/min 550 – 150 ^{o}C 1 K/min

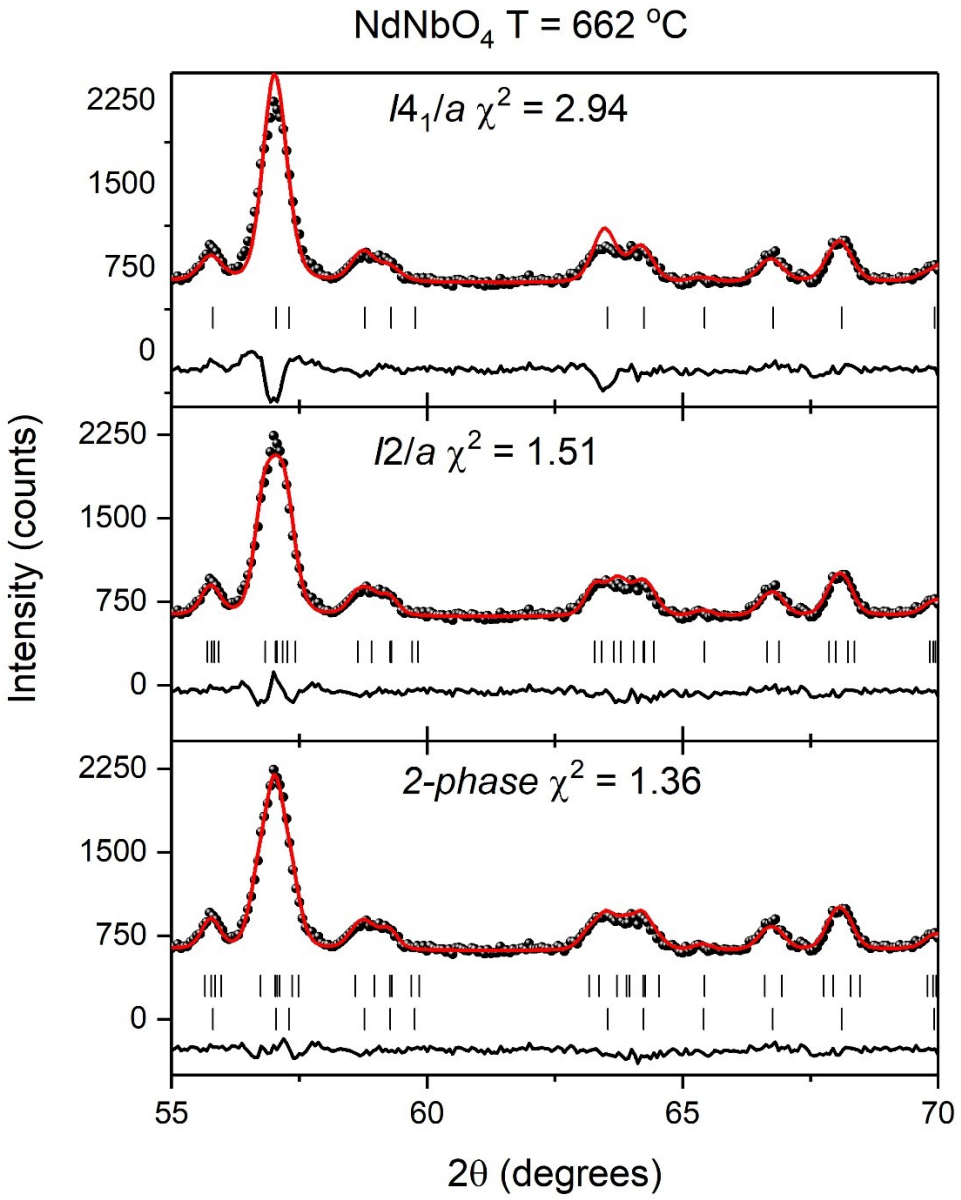


Figure S1. Comparison of the fits obtained for $NdNbO_4$ in the tetragonal $I4_1/a$ (top) monoclinic $I2/a$ (middle) and two phase ($I4_1/a$ and $I2/a$) bottom. In the two phase fit the monoclinic reflections are indicated by the upper tick marks and the tetragonal reflections by the lower tick marks. The refined structural parameters for the three models are listed below.

Monoclinic Only

a = 5.3412(4) b = 11.4203(6) c = 5.3149(4) Å β= 90.337(5)^0 Vol = 324.20(4) Å^3

Atom	X	Y	Z	Ui/Ue*100
Nd1	0.250000	0.1257(9)	0.000000	1.96(7)
Nb1	0.250000	0.6225(11)	0.000000	2.90(10)
O1	0.0020(8)	0.7125(5)	0.1717(8)	3.36(15)
O2	0.9118(8)	0.4613(5)	0.2453(8)	3.66(17)

Tetragonal Only

a = 5.3284(4) c = 11.4200(10) Å Vol = 324.24(4) Å^3

Atom	X	Y	Z	Ui/Ue*100
Nb1	0.000000	0.250000	0.125000	3.00(14)
Nd1	0.000000	0.250000	0.625000	2.04(10)
O1	0.1670(4)	0.0037(6)	0.21184(23)	3.55(8)

Two Phase

a = 5.3468(6) b = 11.4197(21) c = 5.3087(6) Å β = 90.482(10)^0 Vol = 324.13(8) Å^3

Name	X	Y	Z	Ui/Ue*100
Nd1	0.250000	0.1254(10)	0.000000	1.92(6)
Nb1	0.250000	0.6222(13)	0.000000	2.92(9)
O1	0.0009(14)	0.7127(13)	0.1751(12)	2.88(23)
O2	0.9115(14)	0.4615(13)	0.2452(14)	3.24(25)

a = 5.3279(4) c = 11.4217(30) Å Volume = 324.22(9) Å^3

Name	X	Y	Z	Ui/Ue*100
Nd	0.000000	0.250000	0.625000	1.92(6)
Nb	0.000000	0.250000	0.125000	2.92(9)
O1	0.1641(14)	0.0035(16)	0.2115(19)	4.05(25)

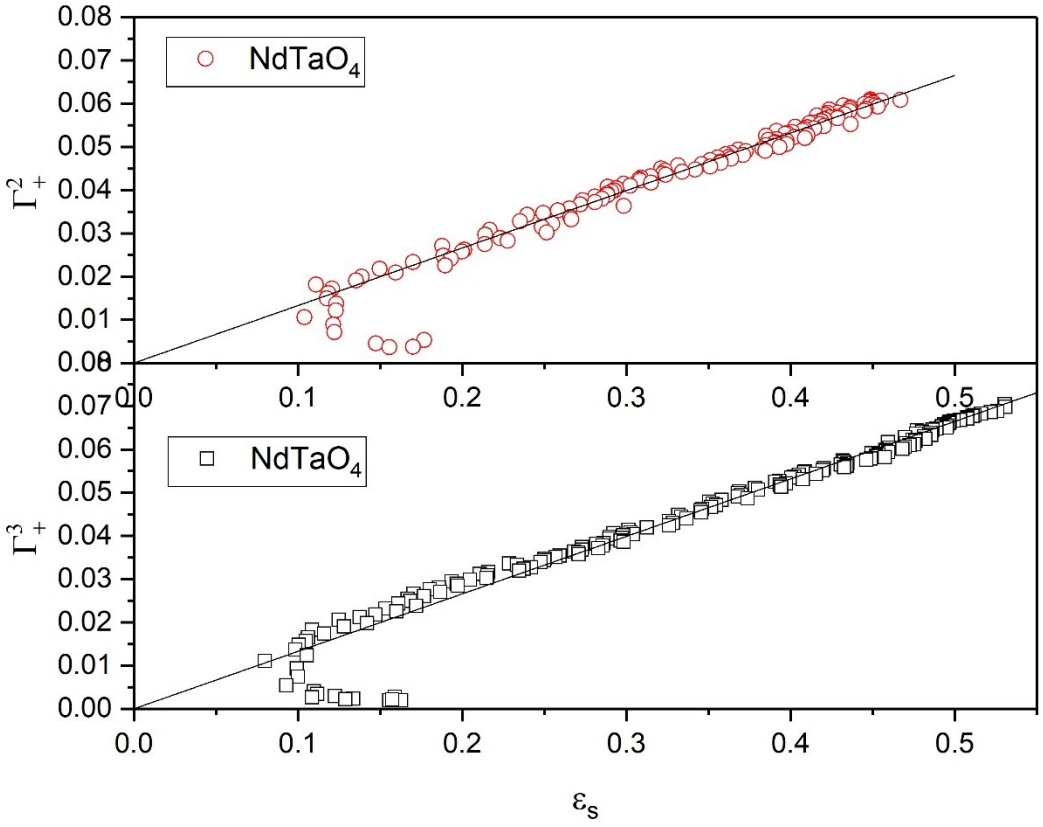


Figure S2. plots of $\Gamma_2^+ \; vs \; \varepsilon^s$ for $NdNbO_4$ and $NdTaO_4$. The solid line is a linear fit to the data with the intercept fixed to be the origin.

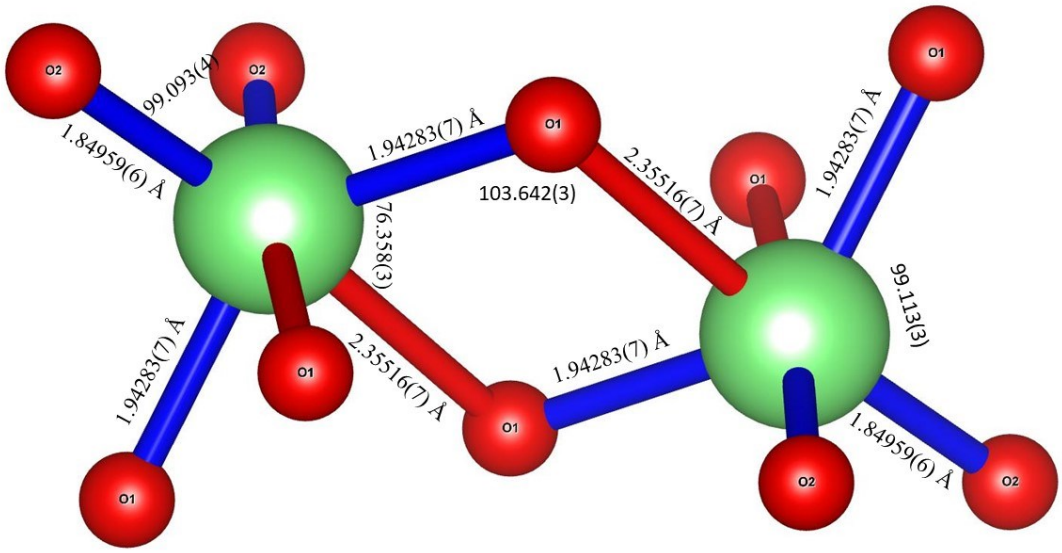


Figure S3 Representation of the local environment in monoclinic $NdNbO_4$. The large Green spheres represent the Nb cations. The four short Nb-O bonds are illustrated by the blue lines and the two long Nb-O bonds by the red lines.

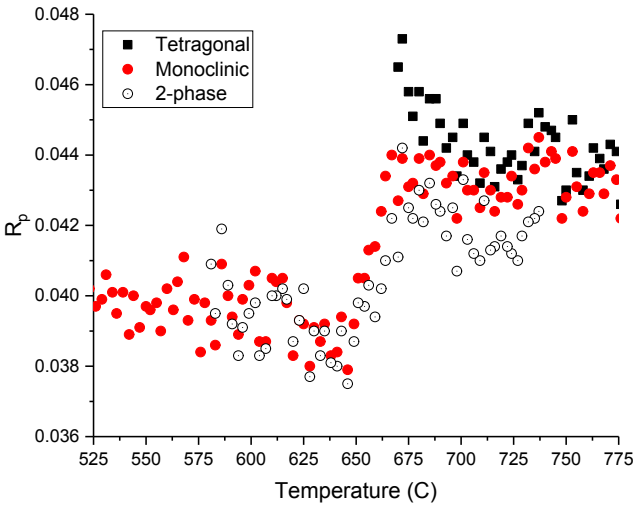
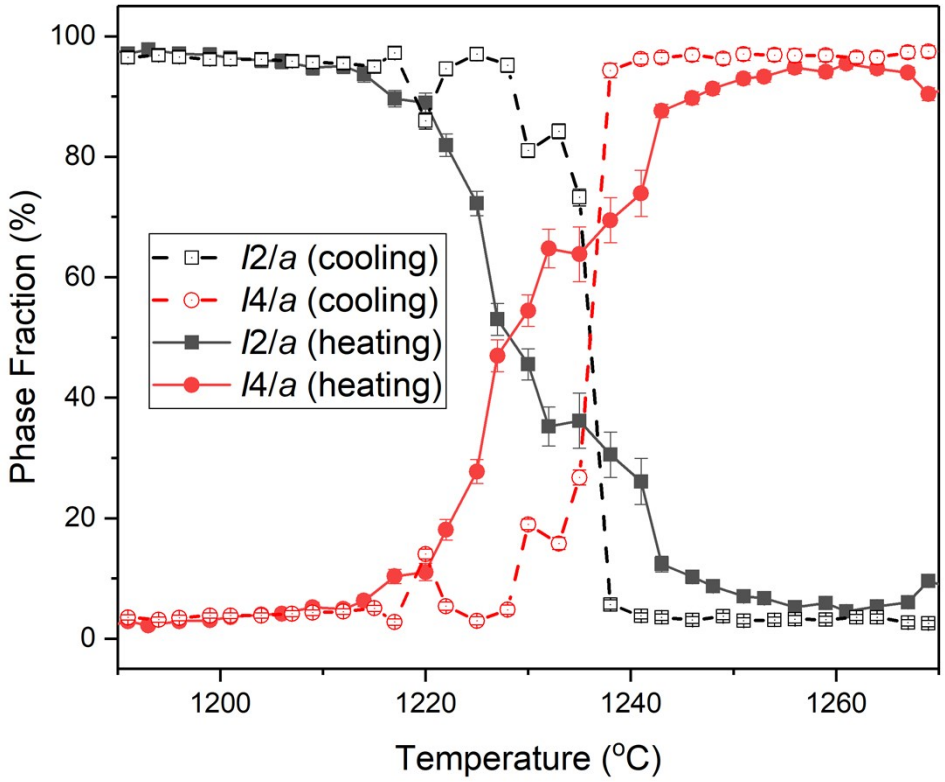


Figure S4. R-factors across the two phase region in $NdNbO_4$ for the data collected on heating.



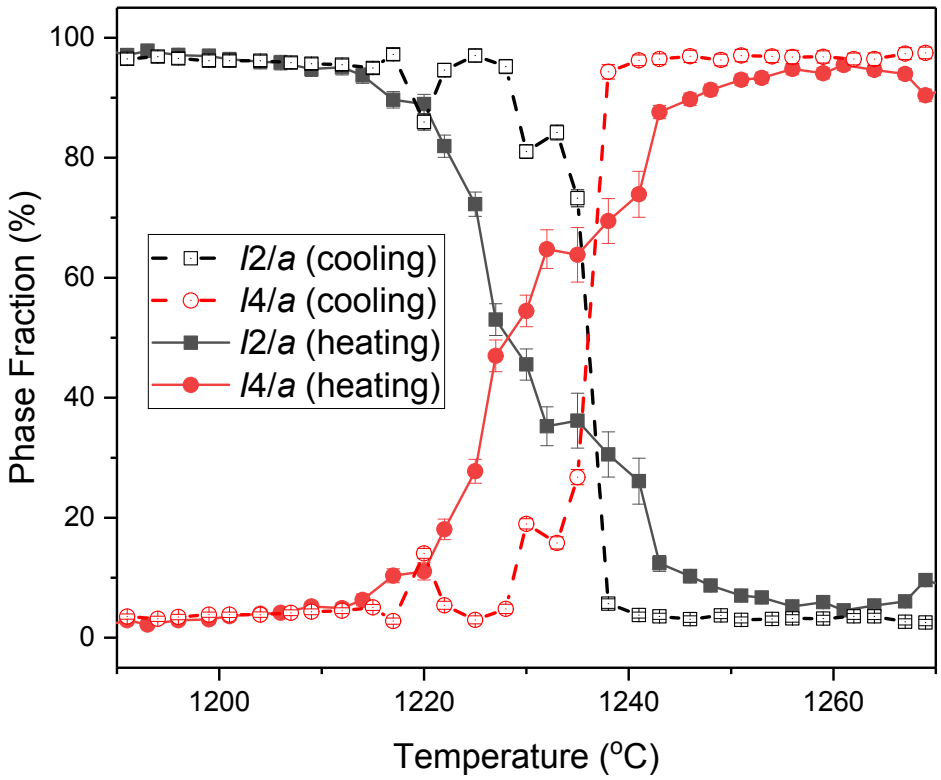


Figure S5. Temperature dependence of the tetragonal and monoclinic phase fractions in $NdTaO_4$ upon heating and cooling. The observed hysteresis and phase coexistence is suggestive of a first order transition. Where not apparent the esds are smaller than the symbols