

Electronic Supporting Information for:

Comparative study of the magnetic properties of $\text{La}_3\text{Ni}_2B'\text{O}_9$ for $B' = \text{Nb}, \text{Ta}$ or Sb

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Table S1. Structural parameters of $\text{La}_3\text{Ni}_2\text{NbO}_9$ at 50 K.

| Atom | Site | x | y | z | $U_{\text{iso}}/\text{\AA}^2$ | Occupancy |
|--------|------|---------------|---------------|------------|-------------------------------|------------------------------|
| La | $4e$ | 0.4916(5) | 0.4585(2) | 0.2504(12) | 0.0145(3) | 1 |
| Ni/Nb1 | $2c$ | 0 | $\frac{1}{2}$ | 0 | 0.0017(2) | Ni: 0.964(7) Nb: 0.036(7) |
| Ni/Nb2 | $2d$ | $\frac{1}{2}$ | 0 | 0 | 0.0017(2) | Ni: 0.369(7) Nb: 0.631(7) |
| O1 | $4e$ | 0.7915(11) | 0.7978(9) | -0.0380(7) | 0.0019(12) | 1 |
| O2 | $4e$ | 0.7111(12) | 0.2828(10) | -0.0468(7) | 0.0074(14) | 1 |
| O3 | $4e$ | 0.5779(4) | 0.0174(4) | 0.2511(14) | 0.0094(5) | 1 |

$R_{\text{wp}} = 4.71\%$, $R_{\text{p}} = 3.62\%$, $\chi^2 = 4.598$

Space group $P2_1/n$: $a = 5.5757(1)\text{ \AA}$, $b = 5.6382(1)\text{ \AA}$, $c = 7.9033(2)\text{ \AA}$, $\beta = 90.027(8)^\circ$

Table S2. Structural parameters of $\text{La}_3\text{Ni}_2\text{TaO}_9$ at 120 K.

| Atom | Site | x | y | z | $U_{\text{iso}}/\text{\AA}^2$ | Occupancy |
|--------|------|---------------|---------------|------------|-------------------------------|------------------------------|
| La | $4e$ | 0.4917(5) | 0.4597(2) | 0.2492(13) | 0.0151(3) | 1 |
| Ni/Ta1 | $2c$ | 0 | $\frac{1}{2}$ | 0 | 0.0014(2) | Ni: 0.982(1) Ta: 0.018(1) |
| Ni/Ta2 | $2d$ | $\frac{1}{2}$ | 0 | 0 | 0.0014(2) | Ni: 0.351(1) Ta: 0.649(1) |
| O1 | $4e$ | 0.7890(11) | 0.7974(10) | -0.0394(8) | 0.0029(14) | 1 |
| O2 | $4e$ | 0.7081(12) | 0.2833(10) | -0.0452(9) | 0.0066(16) | 1 |
| O3 | $4e$ | 0.5776(5) | 0.0179(4) | 0.2505(11) | 0.0092(5) | 1 |

$R_{\text{wp}} = 4.45\%$, $R_{\text{p}} = 3.48\%$, $\chi^2 = 3.150$

Space group $P2_1/n$: $a = 5.5804(2)\text{ \AA}$, $b = 5.6371(2)\text{ \AA}$, $c = 7.9094(2)\text{ \AA}$, $\beta = 90.064(5)^\circ$

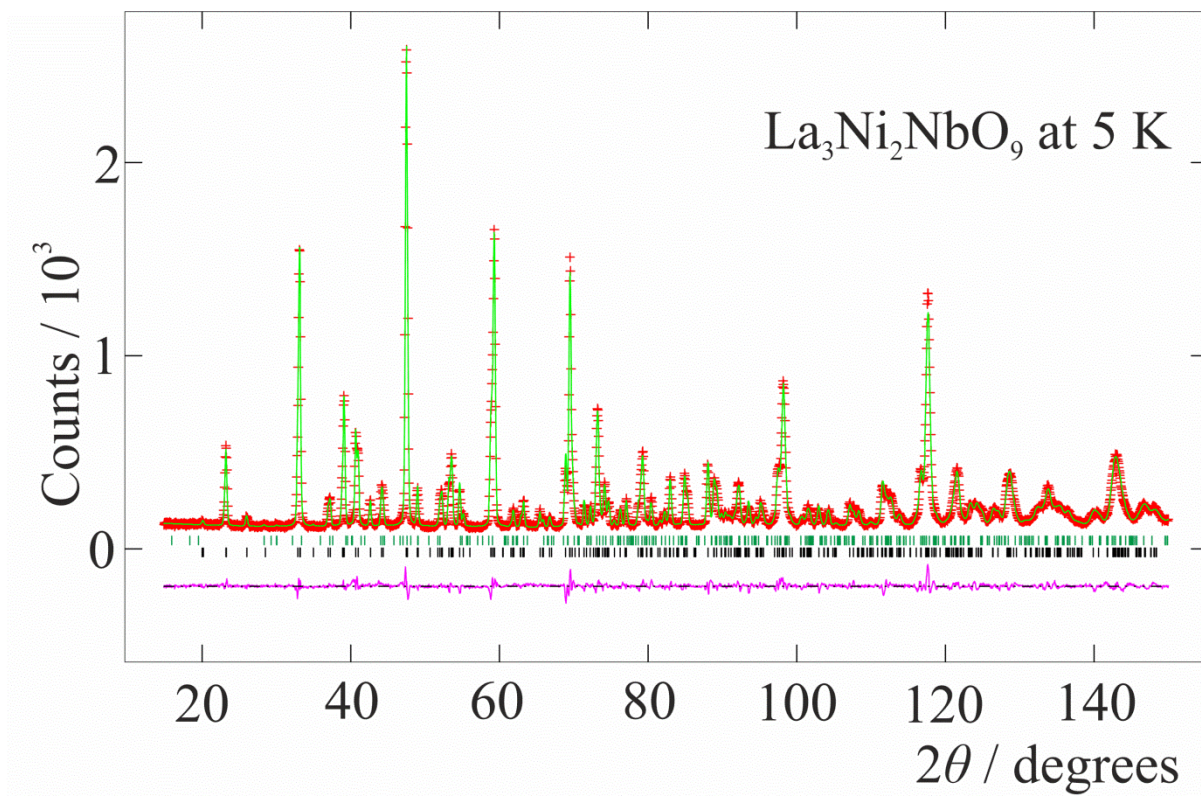
Table S3. Selected bond lengths (Å) and angles (°) in $\text{La}_3\text{Ni}_2B'\text{O}_9$ ($B' = \text{Nb}$ or Ta).

| | $\text{La}_3\text{Ni}_2\text{NbO}_9$ (50 K) | $\text{La}_3\text{Ni}_2\text{TaO}_9$ (120 K) |
|------------------------------|---|--|
| La–O1 | 2.729(9) | 2.756(11) |
| La–O1 | 2.720(8) | 2.702(9) |
| La–O1 | 2.435(8) | 2.440(10) |
| La–O2 | 2.828(10) | 2.806(12) |
| La–O2 | 2.448(8) | 2.437(9) |
| La–O2 | 2.621(8) | 2.651(10) |
| La–O3 | 2.534(2) | 2.536(3) |
| La–O3 | 2.423(3) | 2.426(4) |
| $2c - \text{O1}$ | 2.064(5)×2 | 2.072(5)×2 |
| $2c - \text{O2}$ | 2.057(6)×2 | 2.067(5)×2 |
| $2c - \text{O3}$ | 2.017(11)×2 | 2.024(9)×2 |
| $2d - \text{O1}$ | 2.008(6)×2 | 2.001(5)×2 |
| $2d - \text{O2}$ | 2.016(6)×2 | 2.007(6)×2 |
| $2d - \text{O3}$ | 2.033(11)×2 | 2.030(9)×2 |
| $\text{O1} - 2c - \text{O2}$ | 89.0(3)×2 | 89.7(4)×2 |
| $\text{O1} - 2c - \text{O3}$ | 88.9(2)×2 | 89.1(2)×2 |
| $\text{O2} - 2c - \text{O3}$ | 88.0(2)×2 | 88.4(2)×2 |
| $\text{O1} - 2d - \text{O2}$ | 87.1(3)×2 | 87.7(4)×2 |
| $\text{O1} - 2d - \text{O3}$ | 90.0(2)×2 | 89.5(2)×2 |
| $\text{O2} - 2d - \text{O3}$ | 89.1(2)×2 | 89.3(2)×2 |
| $2c - \text{O1} - 2d$ | 153.6(3) | 153.7(4) |
| $2c - \text{O2} - 2d$ | 153.5(3) | 153.6(4) |
| $2c - \text{O3} - 2d$ | 154.6(1) | 154.6(1) |

Figure Captions

- Figure S1 Observed (red crosses), calculated (green line) and difference (purple line) NPD profiles of (a) $\text{La}_3\text{Ni}_2\text{NbO}_9$ and (b) $\text{La}_3\text{Ni}_2\text{TaO}_9$ at 5 K measured on D2b. The meaning of the reflection markers is the same as in Figures 1 and 2.
- Figure S2 Observed (red crosses), calculated (green line) and difference (purple) neutron powder diffraction profiles of $\text{La}_3\text{Ni}_2\text{NbO}_9$ recorded on D1b at room temperature, 50 K and 5 K. Underneath is a difference curve, purple in colour. The green and cyan vertical markers indicate reflection positions for LaNbO_4 and $\text{La}_3\text{Ni}_2\text{NbO}_9$.
- Figure S3 Observed (red crosses), calculated (green line) and difference (purple line) neutron powder diffraction profiles of $\text{La}_3\text{Ni}_2\text{TaO}_9$ recorded on D1b at 120 and 20 K. Reflection markers for La_3TaO_7 and $\text{La}_3\text{Ni}_2\text{TaO}_9$ are cyan and black respectively.
- Figure S4 Scanning electron microscope (SEM) images of the particles of the $\text{La}_3\text{Ni}_2\text{NbO}_9$ sample (left) and the $\text{La}_3\text{Ni}_2\text{TaO}_9$ sample (right) taken at a voltage of 20kV.
- Figure S5 Particle size distribution (PSD) for $\text{La}_3\text{Ni}_2\text{NbO}_9$ (top) and $\text{La}_3\text{Ni}_2\text{TaO}_9$ (bottom), where the diameter of each particle is calculated from the mean diameter (left) and from the estimated area (right). The mean particle size (μ), standard deviation (σ) and amount of investigated particles (N) are given for each particle size distribution.

(a)



(b)

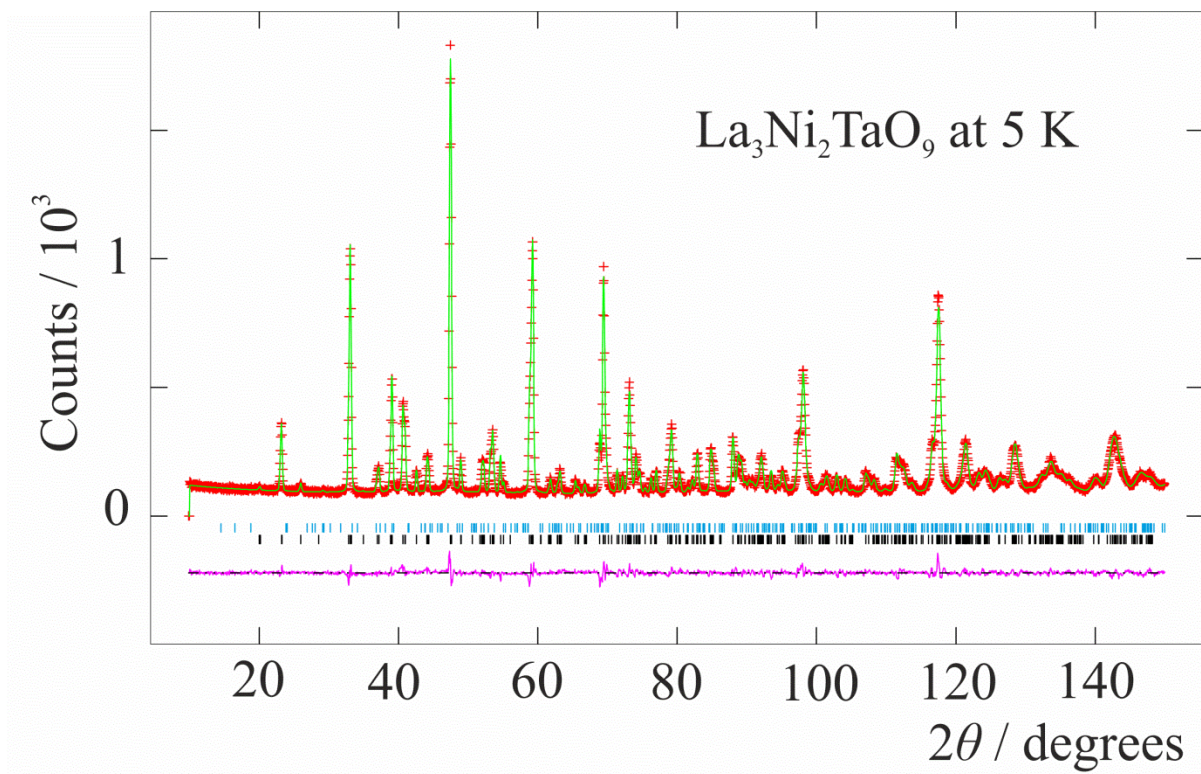


Figure S1

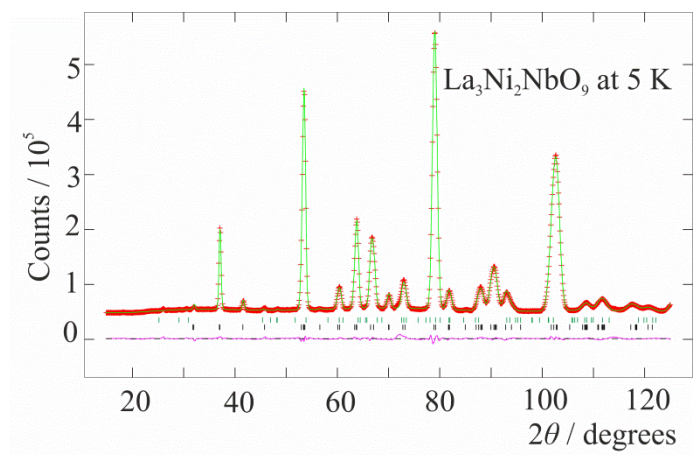
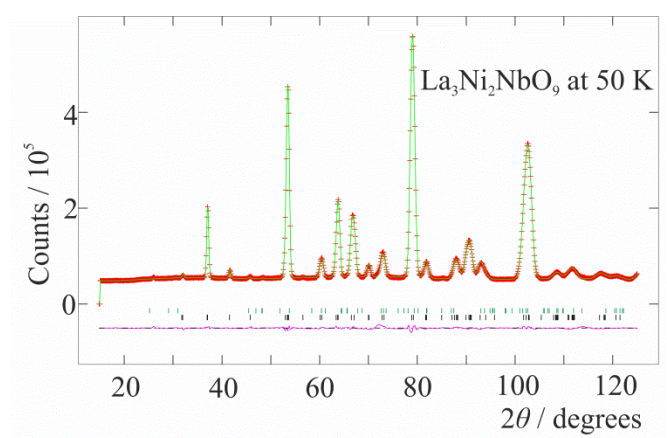
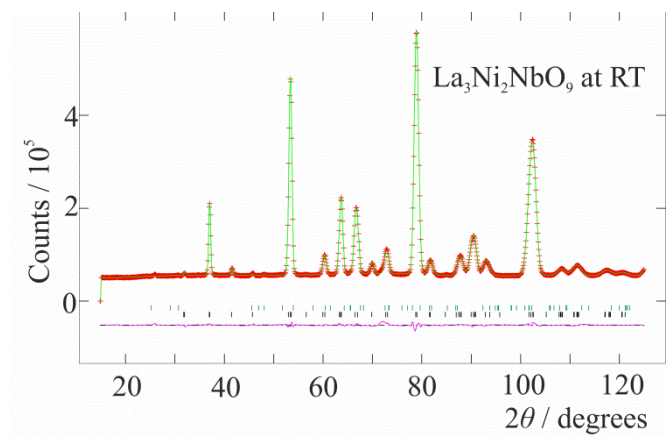


Figure S2

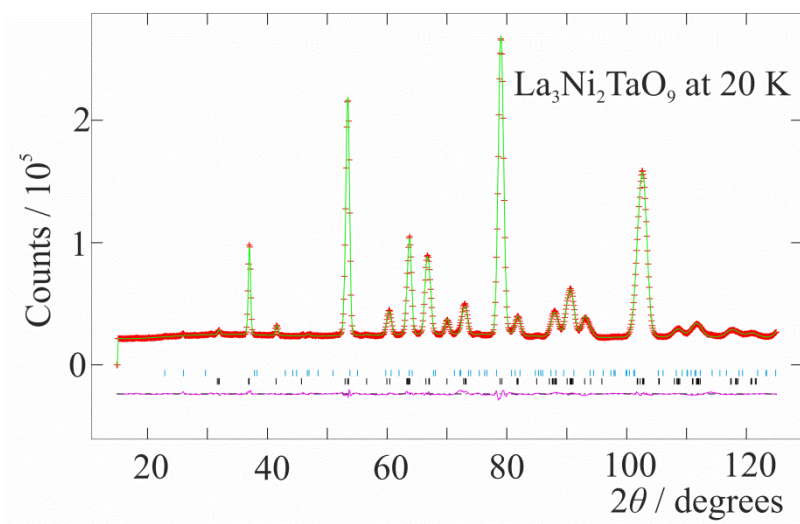
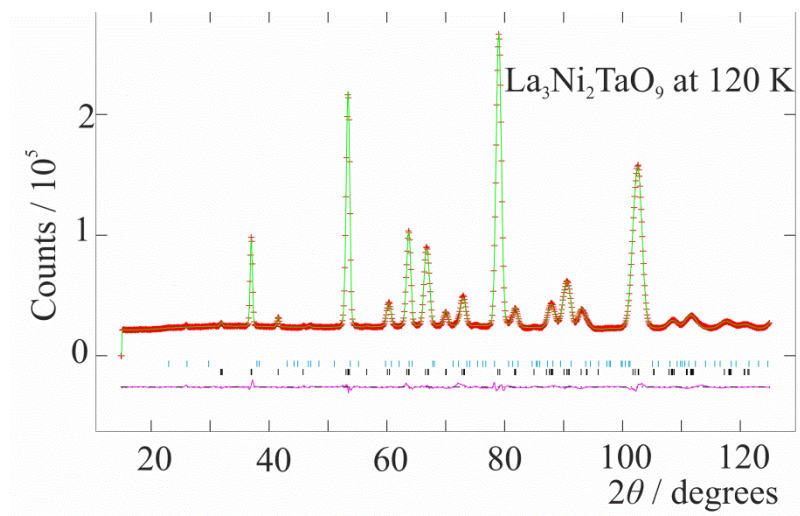


Figure S3

Figure S4

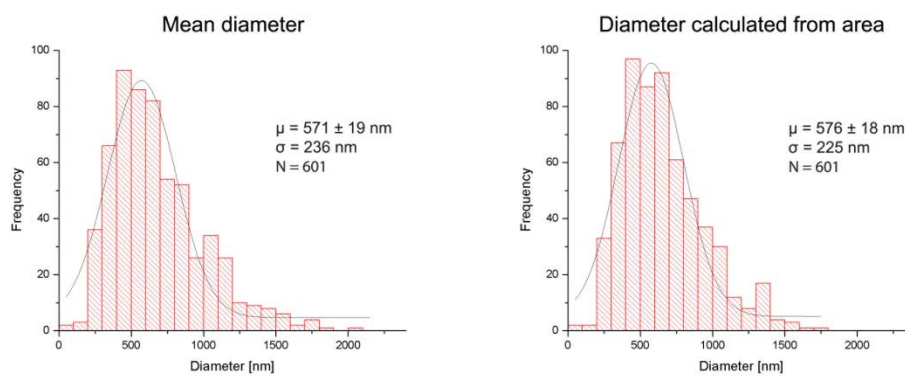
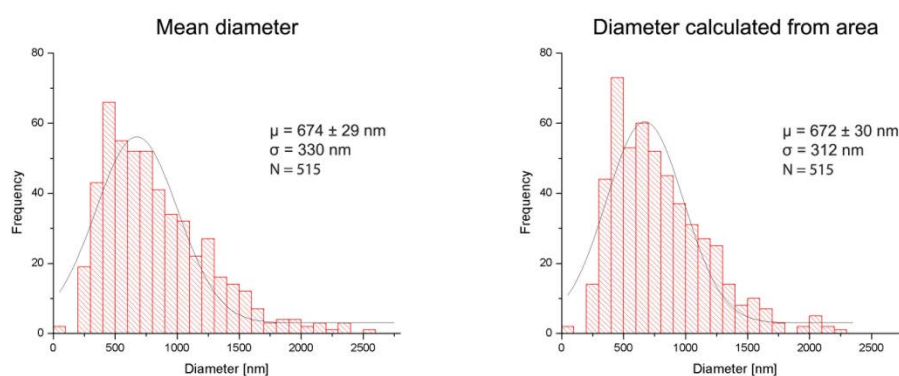


Figure S5