Supporting Information

Spin-degree Manipulation for One-dimensional Room-temperature Ferromagnetism in Haldane System

Pengfei Tan,[a] Chuanhui Zhu,[a] Xiaosheng Ni,[b] Han-Qing Wu,[b] Shuang Zhao,[a] Tao Xia,[a] Jinjin Yang,[a] Tao Han,[a] Mei-Huan Zhao,[a] Yifeng Han,[a] Yuanhua Xia,[c] Zheng Deng,[d] Muwei Wu,[b] Dao-Xin Yao,[b,*] Man-Rong Li [a,e,*]

[a] *Key Laboratory of Bioinorganic and Synthetic Chemistry of Ministry of Education, School of Chemistry, Sun Yat-Sen University, Guangzhou510006, P. R. China*

[b] *Guangdong Key Laboratory of Magnetoelectric Physics and Devices, School of Physics, Sun Yat-Sen University, Guangzhou 510275, P. R. China*

[c] *Key Laboratory of Neutron Physics and Institute of Nuclear Physics and Chemistry, China Academy of Engineering Physics (CAEP), Mianyang 621999, P. R. China*

[d] *Institute of Physics, Chinese Academy of Sciences, Beijing 100190, P. R. China*

[e] *School of Science, Hainan University, Haikou 570228, P. R. China*

E-mail: limanrong@mail.sysu.edu.cn, yaodaox@mail.sysu.edu.cn

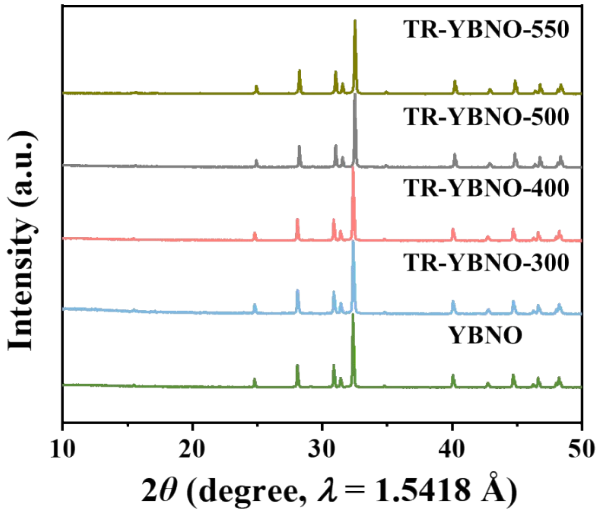

Fig. S1 PXD patterns for YBNO before and after TR reactions under different temperatures.

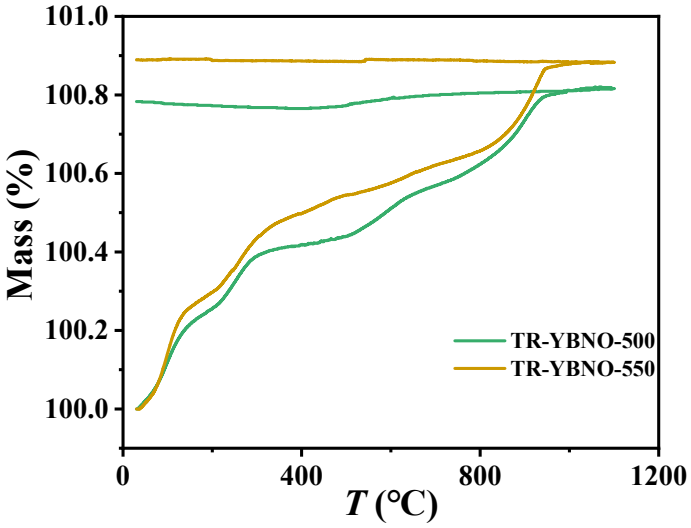

Fig. S2 TGA measurements of TR-YBNO-500 and TR-YBNO-550 up to 1100 °C in air.

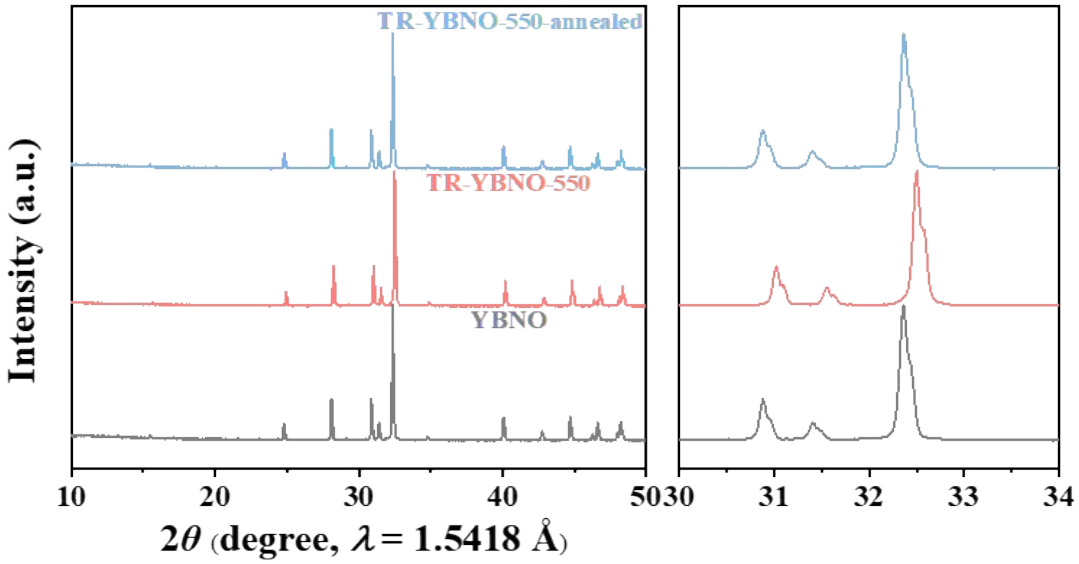

Fig. S3 PXD patterns for YBNO, TR-YBNO-550 and TR-YBNO-550 annealed in air for 800 °C.

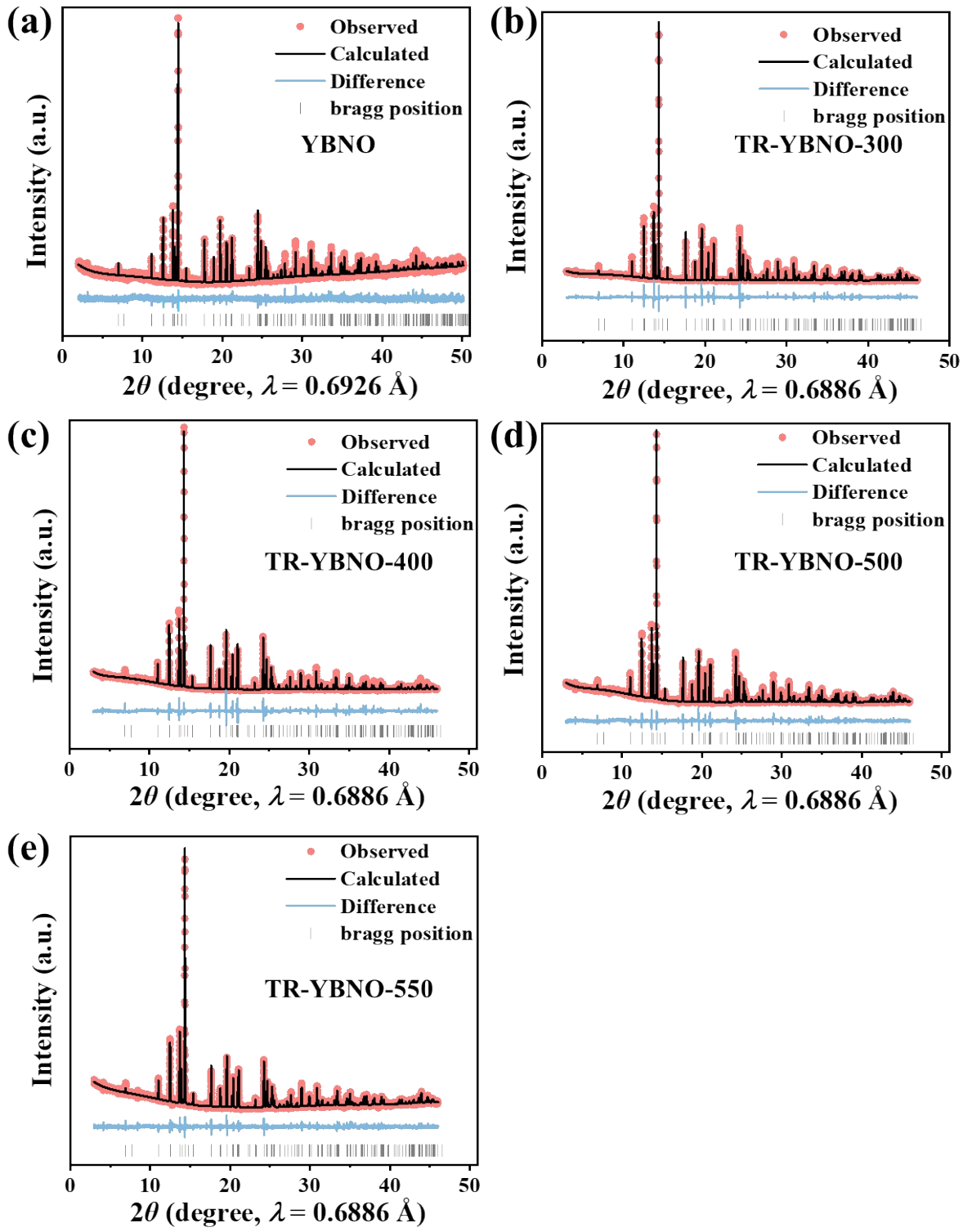

Fig. S4 Refinements of the SPXD data of YBNO and TR-YBNO under different TR temperatures. Experimental, calculated, difference curves and peak positions are shown as red, black, blue, and gray, respectively.

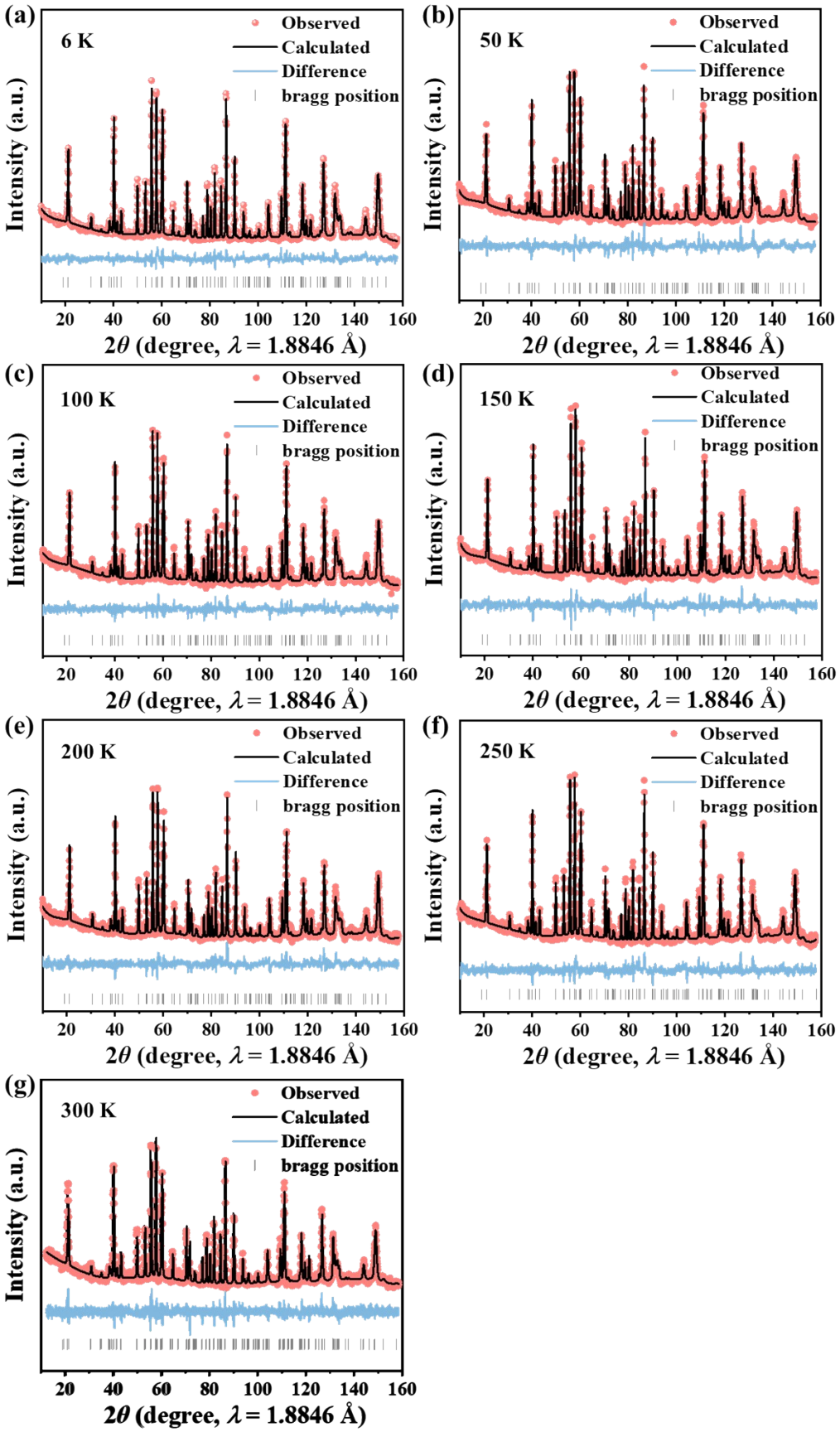

Fig. S5 Refinements of the *in situ* VT-NPD data of TR-YBNO-550. Experimental, calculated, difference curves and peak positions are shown as red, black, blue, and gray, respectively.

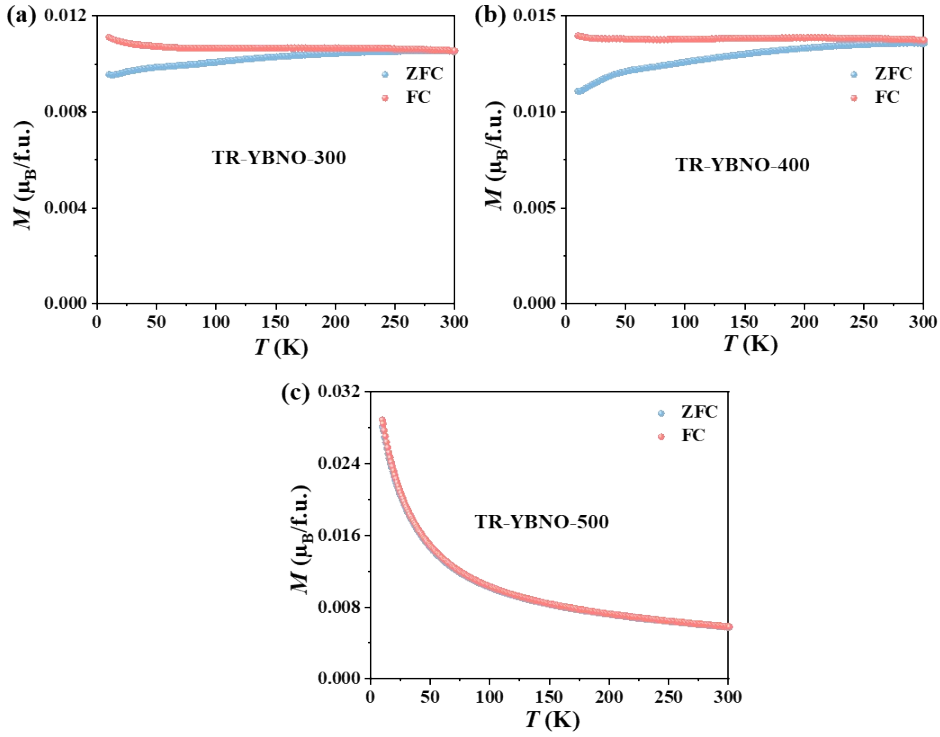

Fig. S6 Temperature-dependent magnetization curves TR-YBNO under different TR reaction temperature.

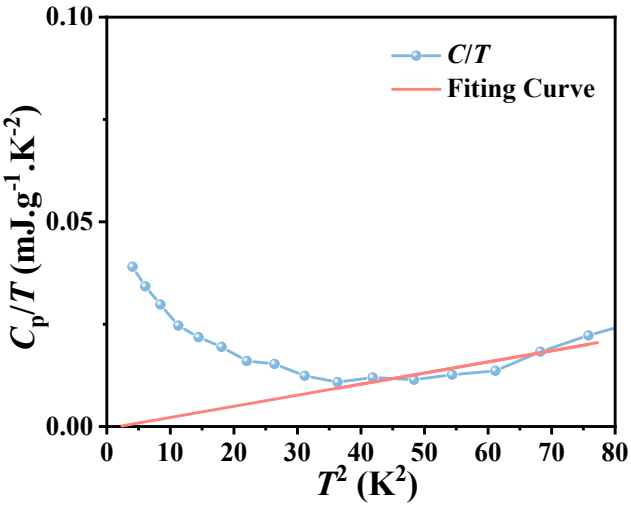

Fig. S7 Fitting curve of *C*/*T vs.* T^2 data for TR-YBNO-550 using a linear model.

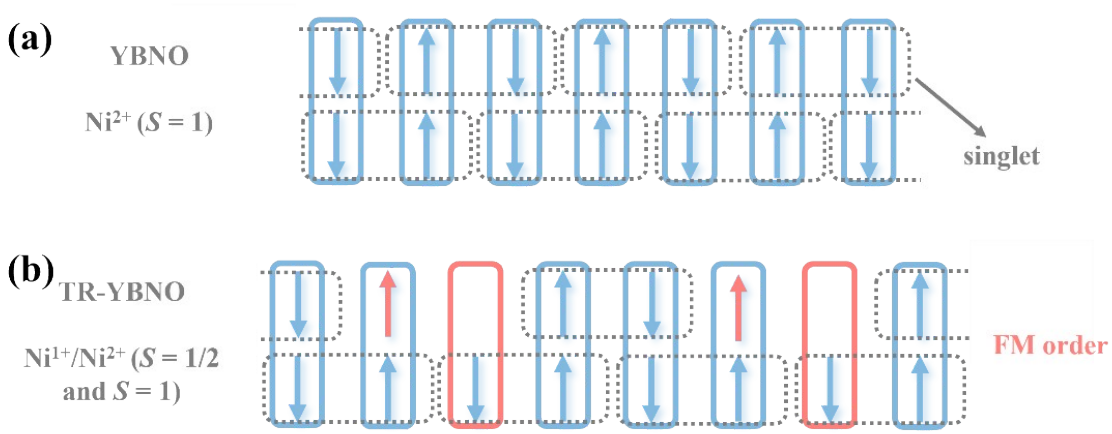

Fig. S8 (a) VBS state in $S = 1$ 1D AFM chain. (b) Interaction between two end $S = 1/2$ spins through the finite Haldane chain with odd number of sites.

Supplementary Tables

Table S1. Crystal structure parameters of the Rietveld refinements of the SPXD data of the TR-YBNO -300, TR-YBNO -400, TR-YBNO-500, TR-YBNO-550.

Sample	TR-YBNO-300	TR-YBNO-400	TR-YBNO-500	TR-YBNO-550
a (Å)	3.7621(4)	3.7617(1)	3.7600(1)	3.7584(5)
b (Å)	5.7630(5)	5.7624(3)	5.7622(2)	5.76198(8)
c (Å)	11.3354(2)	11.3350(1)	11.3338(2)	11.3332(2)
V (Å^3)	245.75(2)	245.70(3)	245.56(5)	245.43(1)
Ba1 (*x*, *y*, *z*)	0, 0, 0	0, 0, 0	0, 0, 0	0, 0, 0
B_{iso}, Å^2	0.65(1)	0.59(5)	0.50(3)	0.67(1)
Ni1 (*x*, *y*, *z*)	0.5, 0.5, 0	0.5, 0.5, 0	0.5, 0.5, 0	0.5, 0.5, 0
B_{iso}, Å^2	0.49(2)	0.42(4)	0.32(8)	0.28(4)
Y1 (*x*, *y*, *z*)	0.5, 0, 0.7029(1)	0.5, 0, 7027(2)	0.5, 0, 7029(6)	0.5, 0, 0.7025(2)
B_{iso}, Å^2	0.52(4)	0.50(3)	0.32(5)	0.37(2)
O1 (*x*, *y*, *z*)	0.5, 0, 0.5	0.5, 0, 0.5	0.5, 0, 0.5	0.5, 0, 0.5
B_{iso}, Å^2	0.47(1)	0.24(4)	0.21(5)	0.18 (5)
Occ	0.99(2)	0.97(2)	0.96(2)	0.94 (1)
O2 (*x*, *y*, *z*)	0, 0.7544(2), 0.3421(5)	0, 0.7510(2), 0.3415(3)	0, 0.7546(2), 0.3409(6)	0, 0.7525(2), 0.3401(5)
B_{iso}, Å^2	0.48(2)	0.44(3)	0.15(5)	0.40(4)
Occ	0.99(1)	0.99(3)	0.97(1)	0.96(1)
R_{wp}	4.87	4.03	4.30	2.44
R_p	2.81	2.43	2.57	1.74

Table S2. Crystal structure parameters of the Rietveld refinements of the in situ variable temperature NPD data of the as-made TR-YBNO-550 ($Y_2BaNiO_{4.75}$) between 6 and 300 K (orthorhombic, space group *Immm* (No. 71))

T/K	6	50	100	150	200	250	300
a (Å)	3.7548(1)	3.7550(1)	3.7555(1)	3.7566(1)	3.7579(1)	3.7593(1)	3.7606(1)
b (Å)	5.7502(1)	5.7508(2)	5.7522(2)	5.7553(2)	5.7585(2)	5.7616(2)	5.7649(2)
c (Å)	11.3192(2)	11.3198(3)	11.3224(3)	11.3267(3)	11.3315(3)	11.3370(3)	11.3418(3)
V (Å^3)	244.39(1)	244.45(1)	244.59(1)	244.89(1)	245.21(1)	245.55(1)	245.88 (1)
Ba1 (*x*, *y*, *z*)	0, 0, 0	0, 0, 0	0, 0, 0	0, 0, 0	0, 0, 0	0, 0, 0	0, 0, 0
B_{iso}, Å^2	0.51(4)	0.54(1)	0.64(4)	0.71(5)	0.81(3)	0.98(1)	1.16(1)
Ni1 (*x*, *y*, *z*)	0.5, 0.5, 0	0.5, 0.5, 0	0.5, 0.5, 0	0.5, 0.5, 0	0.5, 0.5, 0	0.5, 0.5, 0	0.5, 0.5, 0
B_{iso}, Å^2	0.32(2)	0.40(7)	0.47(1)	0.58(6)	0.66(3)	0.72(6)	0.79(2)
Y1 (*x*, *y*, *z*)	0.5, 0, 0.7038(2)	0.5, 0, 0.7040(4)	0.5, 0, 0.7036(3)	0.5, 0, 0.7036(3)	0.5, 0, 0.7037(3)	0.5, 0, 0.7041(3)	0.5, 0, 0.7041(3)
B_{iso}, Å^2	0.42(5)	0.46(1)	0.45(2)	0.61(1)	0.70(3)	0.79(1)	0.86(5)
O1 (*x*, *y*, *z*)	0.5, 0, 0.5	0.5, 0, 0.5	0.5, 0, 0.5	0.5, 0, 0.5	0.5, 0, 0.5	0.5, 0, 0.5	0.5, 0, 0.5
B_{iso}, Å^2	0.28(1)	0.51(3)	0.62(3)	0.66(5)	0.73(4)	0.80(2)	0.96(2)
Occ	0.94(1)	0.94(1)	0.94(1)	0.94(1)	0.94(1)	0.94(1)	0.94(1)
O2 (*x*, *y*, *z*)	0, 0.7584(9), 0.3451(2)	0, 0.7577(2), 0.3432(3)	0, 0.7578(1), 0.3445(5)	0, 0.7575(2), 0.3448(1)	0, 0.7570(1), 0.3445(3)	0, 0.7594(1), 0.3441(4)	0, 0.7573(1), 0.3435(3)
B_{iso}, Å^2	0.24(2)	0.41(4)	0.42(7)	0.45(2)	0.55(4)	0.66(1)	0.74(5)
Occ	0.96(1)	0.96(1)	0.96(1)	0.96(1)	0.96(1)	0.96(1)	0.96(1)
R_{wp}	5.47	7.56	6.49	6.86	7.02	6.79	6.51
R_p	4.26	5.81	4.93	5.26	5.48	5.26	5.00

Table S3. The DFT calculated Heisenberg exchange interaction parameters and the corresponding bond length. The single-ion anisotropy constant D of Eq. (1) is also listed.

	S^2J_1	S^2J_2	S^2J_3	S^2D
Bond (Å)	3.76	5.76	6.63	-
Value (meV)	57.08	3.74	0.35	0.02