SUPPLEMENTAL MATERIAL: High-pressure synthesis of quantum magnet M-$YbTaO_4$ with a stretched diamond lattice

Nicola D. Kelly,[1,2,*] Xuan Liang,[2] Siân E. Dutton,[1,†]
Kazunari Yamaura,[2] and Yoshihiro Tsujimoto[2,‡]

[1] *Cavendish Laboratory, University of Cambridge, J J Thomson Avenue, Cambridge, CB3 0US, United Kingdom*

[2] *Research Center for Materials Nanoarchitectonics (MANA), National Institute for Materials Science (NIMS), 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan*

* ne281@cam.ac.uk

† sed33@cam.ac.uk

‡ tsujimoto.yoshihiro@nims.go.jp

TABLE S1. All synthesis attempts to form $YbTaO_4$. All samples were prepared from the constituent oxides (Yb_2O_3 and Ta_2O_5), *except* the samples marked with a dagger(†), which were prepared from M'-$YbTaO_4$ precursor (NK067) pre-synthesised in a furnace at ambient pressure.

Sample	P (GPa)	T (°C)	Time (h)	M (%)	M' (%)	Other
NK067	Ambient	1500	24	0	100	–
NK068	Ambient	1500	24	0	100	–
NK053	6	1200	4	0	0	Yb_3TaO_7 + Ta_2O_5
NK058	6	1400	1	2	98	–
NK063	6	1500	1	5	95	–
NK059	6	1600	2	50	50	–
NK064	6	1700	1	97	1	2 % Ta_2O_5
NK070(†)	6	1700	1	85	14	1 % Yb_2O_3
NK071	6	1700	2	95.1	0	4.9 % PtO
NK072(†)	6	1700	2	96.0	3.0	1.0 % Yb_2O_3
NK078	6	1800	1	96.3	0	3.7 % PtO
NK079	6	1800	1	97.1	0	2.9 % PtO
NK080	6	1800	1	95.1	0	4.9 % PtO
NK096	6	1800	1	94.8	0	3.6 % PtO, 1.6 % Ta_2O_5
NK097	6	1800	1	96.7	0	2.8 % PtO, 0.5 % Ta_2O_5
NK098A	6	1800	1	98.0	0	2.0 % PtO
NK099	6	1800	1	97.0	0	3.0 % PtO
NK100	6	1800	1	96.0	0	4.0 % PtO
NK101	6	1800	1	97.7	0	2.3 % PtO
NK102	6	1800	1	98.4	0	1.6 % PtO

TABLE S2. All synthesis attempts to form Nb-doped samples $YbTa_{1-x}Nb_xO_4$. All samples were prepared from the constituent oxides Yb_2O_3, Ta_2O_5 and Nb_2O_5.

Sample	Target x	P (GPa)	T (°C)	Time (h)	M (%)	M' (%)	Other
NK081	0.1	6	1800	1	97.6	0	2.4 % PtO
NK082	0.2	6	1800	1	97.8	0	2.2 % PtO
NK083	0.3	6	1800	1	99.2	0	0.8 % PtO
NK089	0.4	6	1800	1	99.4	0	0.6 % PtO
NK090	0.5	6	1800	1	100	0	–
NK091	0.6	6	1800	1	100	0	–
NK092	0.7	6	1800	1	100	0	–
NK093A	0.8	6	1800	1	100	0	–
NK094	0.9	6	1800	1	100	0	–
NK095	1.0	6	1800	1	100	0	–
NK067	0	Ambient	1500	24	0	100	–
NK103	0.05	Ambient	1500	24	0	100	–
NK084	0.1	Ambient	1500	24	12.2	86.8	–
NK104	0.15	Ambient	1500	24	54.5	45.5	–
NK073	0.2	Ambient	1500	24	94.4	5.6	–
NK085	0.3	Ambient	1500	24	98.3	1.7	–
NK074	0.4	Ambient	1500	24	98.9	1.1	–
NK086	0.5	Ambient	1500	24	100	0	–
NK075	0.6	Ambient	1500	24	100	0	–
NK087	0.7	Ambient	1500	24	100	0	–
NK076	0.8	Ambient	1500	24	100	0	–
NK088	0.9	Ambient	1500	24	100	0	–
NK077	1.0	Ambient	1500	24	100	0	–

TABLE S3. Refined unit cell parameters for M-$YbTaO_4$ samples synthesised at high pressure. Space group $I2/a$, Cu Kα radiation, room temperature.

Sample	a (Å)	b (Å)	c (Å)	β (°)	V (Å^3)	R_{wp} (%)	χ^2
NK071	5.26877(11)	10.8351(2)	5.03043(9)	95.4500(10)	285.877(9)	9.80	2.81
NK078	5.26797(15)	10.8371(3)	5.03117(13)	95.4400(13)	285.933(13)	10.13	2.72
NK079	5.26751(14)	10.8324(3)	5.02981(12)	95.4365(12)	285.709(12)	9.66	2.88
NK080	5.2692(2)	10.8388(4)	5.03159(18)	95.4562(13)	286.061(19)	9.92	2.96
NK096	5.27124(11)	10.8402(2)	5.03350(9)	95.4446(10)	286.322(10)	9.96	2.99
NK097	5.27061(10)	10.83847(19)	5.03253(8)	95.4446(9)	286.188(9)	9.71	2.92
NK098A	5.26978(13)	10.8379(3)	5.03232(11)	95.4480(11)	286.115(12)	10.77	3.56
NK099	5.27048(13)	10.8378(3)	5.03233(11)	95.4462(12)	286.152(12)	10.85	3.63
NK100	5.26896(15)	10.8382(3)	5.03216(12)	95.4534(14)	286.066(13)	10.41	2.92
NK101	5.27149(10)	10.83862(17)	5.03313(8)	95.4507(11)	286.271(8)	11.49	4.09
NK102	5.27046(9)	10.8371(2)	5.03250(9)	95.4466(9)	286.140(9)	10.14	2.89

TABLE S4. Fitted Curie-Weiss parameters for M-$YbTaO_4$ samples using the two-level Curie-Weiss function (Eq. 3 in main text).

Sample	μ_0 (μ_B)	μ_1 (μ_B)	ΔE_{10} (K)	θ_{CW} (K)
NK071	3.55	5.91	262	–2.5
NK078	3.49	5.73	265	–2.8
NK079	3.53	5.78	233	–1.7
NK080	3.62	6.70	287	–3.8
NK096	3.55	5.90	247	–2.0
NK097	3.41	5.58	241	–2.2
NK098A	3.57	5.61	275	–1.6
NK099	3.49	5.62	264	–2.0
NK100	3.40	5.92	262	–3.1
NK101	3.39	5.74	248	–2.3
NK102	3.37	5.56	234	–2.0

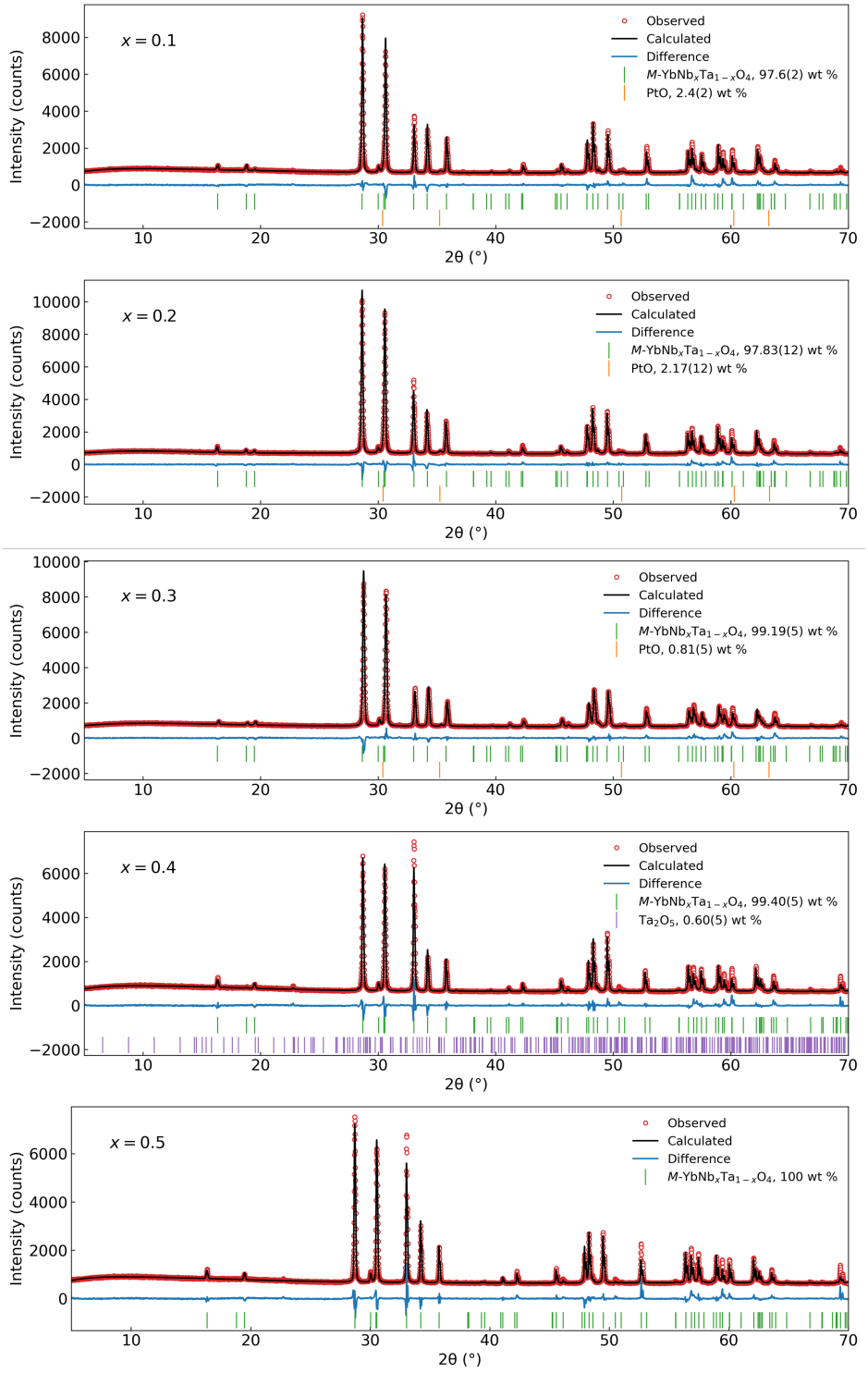


FIG. S1. Rietveld refinements against room-temperature PXRD data (λ = Cu Kα) for $YbNb_xTa_{1-x}O_4$ with $x = 0.1, 0.2, 0.3, 0.4, 0.5$ (top to bottom), synthesised under HPHT conditions (6 GPa, 1800 °C, 1 h). Red circles: observed data, black line: calculated pattern, blue line: difference pattern, tick marks: Bragg reflection positions.

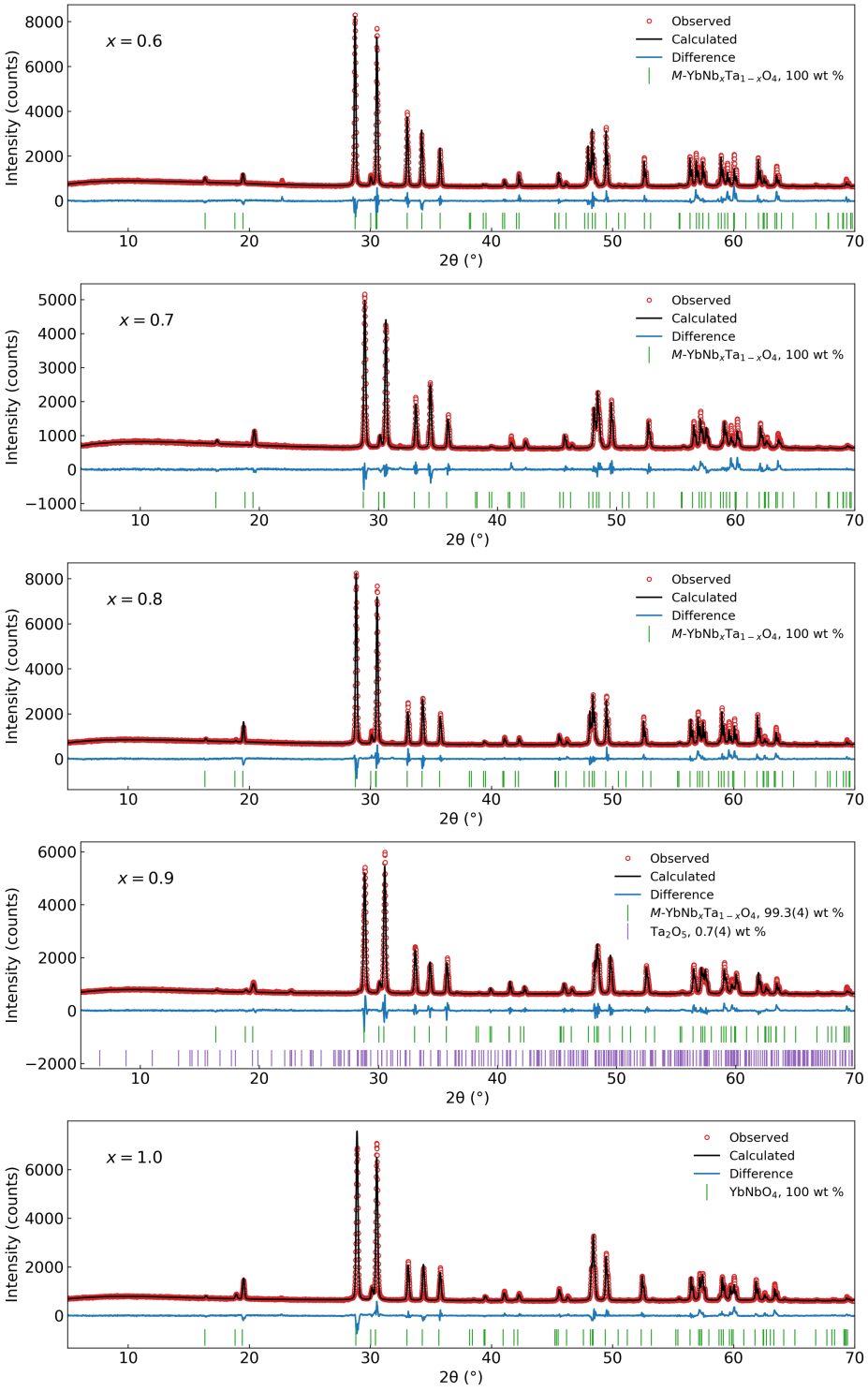


FIG. S2. Rietveld refinements against room-temperature PXRD data (λ = Cu Kα) for $YbNb_xTa_{1-x}O_4$ with $x = 0.6, 0.7, 0.8, 0.9, 1.0$ (top to bottom), synthesised under HPHT conditions (6 GPa, 1800 °C, 1 h), as-made. Red circles: observed data, black line: calculated pattern, blue line: difference pattern, tick marks: Bragg reflection positions.



FIG. S3. Images showing the colour change from off-white or beige to pure white in $YbNb_xTa_{1-x}O_4$ samples after annealing at 800 °C for 2 h.

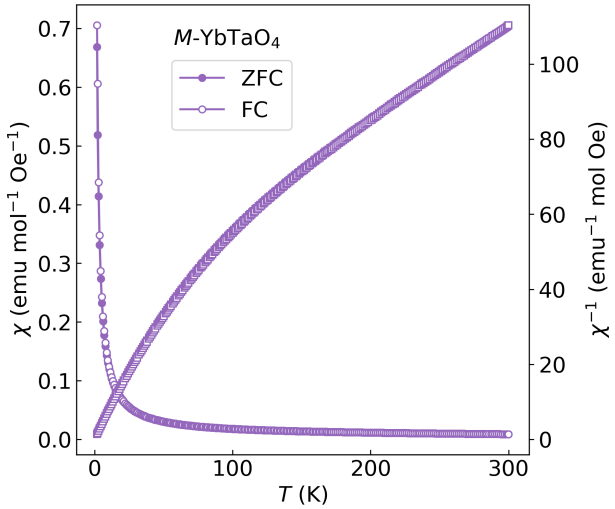


FIG. S4. Magnetic susceptibility $\chi(T)$ and inverse susceptibility $\chi^{-1}(T)$ for a sample of M-$YbTaO_4$ (NK080). The zero-field-cooled (ZFC) and field-cooled (FC) curves match, showing that there is no glassiness.

TABLE S5. Fitted Curie-Weiss parameters for M-$YbTaO_4$ samples using the simple Curie-Weiss law, $\chi = C/(T - \theta_{CW})$, in the high- and low-temperature regimes.

Sample	Range (K)	μ_{eff} (μ_B)	θ_{CW} (K)	Range (K)	μ_{eff}^{LT} (μ_B)	θ_{CW}^{LT} (K)
NK071	150–300	4.96	–87	1.8–30	3.37	–0.63
NK078	150–300	4.82	–86	1.8–30	3.31	–0.87
NK079	150–300	4.83	–70	1.8–30	3.42	–0.74
NK080	150–300	5.61	–135	1.8–30	3.35	–0.88
NK096	150–300	4.94	–79	1.8–30	3.40	–0.63
NK097	150–300	4.68	–75	1.8–30	3.26	–0.66
NK098A	150–300	4.71	–71	1.8–30	3.43	–0.36
NK099	150–300	4.72	–77	1.8–30	3.34	–0.57
NK100	150–300	4.95	–101	1.8–30	3.20	–0.83
NK101	150–300	4.80	–86	1.8–30	3.22	–0.65
NK102	150–300	4.65	–74	1.8–30	3.23	–0.68

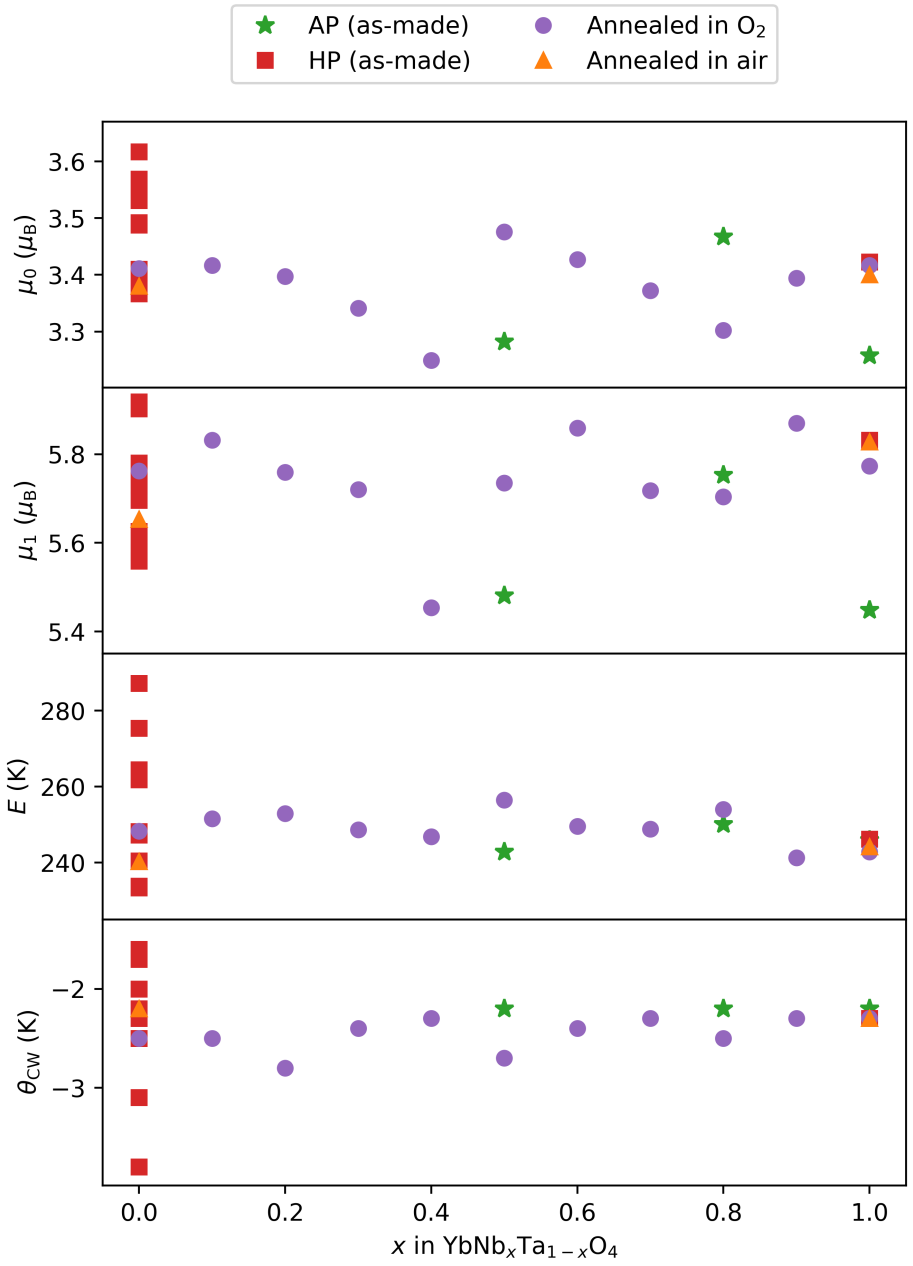


FIG. S5. Fitted magnetic parameters (two-level Curie-Weiss fitting) for selected samples of $YbNb_xTa_{1-x}O_4$.

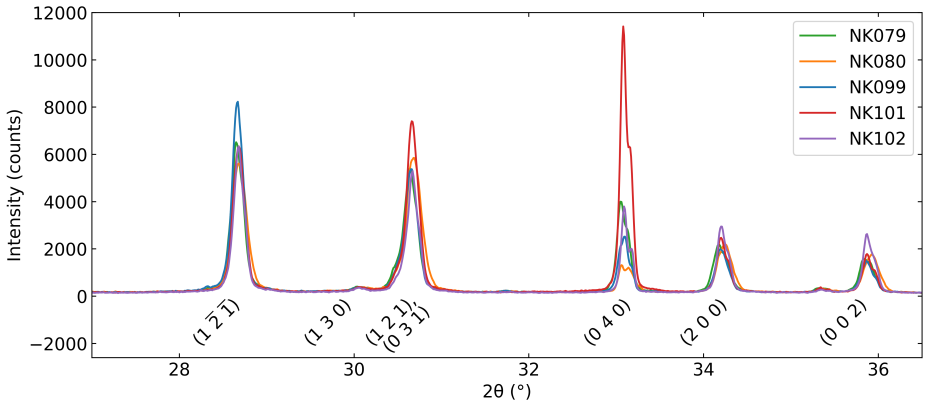


FIG. S6. PXRD data for several samples of $YbTaO_4$ made at high pressure, illustrating the problem of variable preferred orientation, particularly in sample NK101.

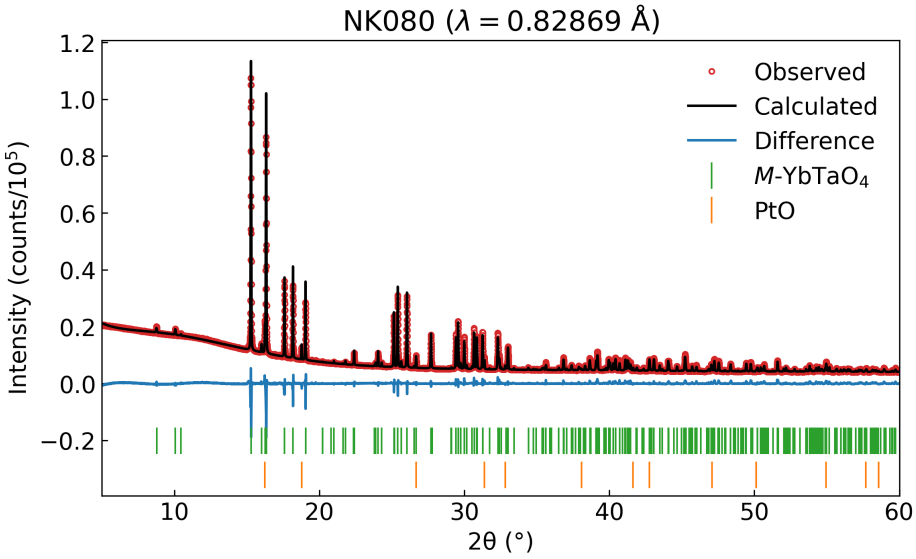


FIG. S7. Synchrotron PXRD data and Rietveld refinement for M-$YbTaO_4$ (high-pressure sample).