# Supplementary materials

**Table S1** Atomic coordinates of $Tb_4Co_2C_3$ refined from NPD ($\lambda$ = 1.28 Å) at 100(1) K and PXRD at 293(2) K ($\lambda$ = 1.54185 Å)

| Atom | Site | x | y | z |
|---|---|---|---|---|
| | | **100 K, $B_{ov}$ = 0.19(5) Å²** | | |
| Tb1 | 2n | 0.2790(6) | 1/2 | 0.9019(12) |
| Tb2 | 2n | 0.7106(6) | 1/2 | 0.6134(12) |
| Tb3 | 2n | 0.0001(8) | 1/2 | 0.7495(13) |
| Tb4 | 2m | 0.4916(5) | 0 | 0.2569(12) |
| Co1 | 2m | 0.8437(13) | 0 | 0.423(4) |
| Co2 | 2m | 0.8564(14) | 0 | 0.929(4) |
| C1 | 2n | 0.1480(5) | 1/2 | 0.5781(14) |
| C2 | 2m | 0.3010(4) | 0 | 0.1459(13) |
| C3 | 1h | 1/2 | 1/2 | 1/2 |
| C4 | 1a | 0 | 0 | 0 |
| | | **293 K, $B_{ov}$ = 0.8(2) Å²** | | |
| Tb1 | 2n | 0.278(1) | 1/2 | 0.901(2) |
| Tb2 | 2n | 0.710(1) | 1/2 | 0.613(2) |
| Tb3 | 2n | 0.000(1) | 1/2 | 0.750(2) |
| Tb4 | 2m | 0.493(5) | 0 | 0.258(2) |
| Co1 | 2m | 0.843(2) | 0 | 0.424(4) |
| Co2 | 2m | 0.856(2) | 0 | 0.927(4) |
| C1 | 2n | 0.148(1) | 1/2 | 0.577(2) |
| C2 | 2m | 0.301(1) | 0 | 0.147(2) |
| C3 | 1h | 1/2 | 1/2 | 1/2 |
| C4 | 1a | 0 | 0 | 0 |

**Table S2** Interatomic distances (in Å) and coordination numbers (CN) for different atom sites in $Tb_4Co_2C_3$ (at 293 K)

| Atom | Neighbor | Distance | CN |
|---|---|---|---|
| Tb1 | – C1 | 2.355(9) | 15 |
| | – C3 (×2) | 2.492(6) | |
| | – Tb1 | 3.40(1) | |
| | – Tb2 (×2) | 3.488(9) | |
| | – Tb1 (×2) | 3.6425(1) | |
| | – Tb3 (×2) | 3.66(1) | |
| | – Tb2 (×2) | 3.70(1) | |
| | – Tb1 | 3.71(1) | |
| | – Tb3 (×2) | 3.876(9) | |
| Tb2 | – C1 (×2) | 2.530(8) | 17 |
| | – C2 | 2.54(1) | |
| | – C3 | 2.600(8) | |
| | – Co1 (×2) | 3.05(2) | |
| | – Co2 (×2) | 3.09(2) | |
| | – Tb3 | 3.41(1) | |
| | – Tb1 (×2) | 3.488(9) | |
| | – Tb4 | 3.57(1) | |
| | – Tb2 (×2) | 3.6425(1) | |
| | – Tb3 | 3.70(1) | |
| | – Tb1 (×2) | 3.70(1) | |
| Tb3 | – C2 | 2.45(1) | 16 |
| | – C1 (×2) | 2.477(8) | |
| | – Co2 (×2) | 2.98(2) | |
| | – Co1 (×2) | 3.03(2) | |
| Tb3 (cont.) | – Tb2 | 3.41(1) | |
| | – Tb4 | 3.44(1) | |
| | – Tb3 (×2) | 3.6425(1) | |
| | – Tb1 (×2) | 3.66(1) | |
| | – Tb2 | 3.70(1) | |
| | – Tb1 (×2) | 3.876(9) | |
| Tb4 | – C2 | 2.51(1) | 18 |
| | – C4 (×2) | 2.546(6) | |
| | – C2 | 2.57(1) | |
| | – Co2 (×2) | 3.10(2) | |
| | – Co2 (×2) | 3.11(2) | |
| | – Co1 (×2) | 3.19(2) | |
| | – Co1 (×2) | 3.19(2) | |
| | – Tb3 | 3.44(1) | |
| | – Tb4 | 3.54(1) | |
| | – Tb4 | 3.56(1) | |
| | – Tb2 | 3.57(1) | |
| | – Tb4 (×2) | 3.6425(1) | |
| Co1 | – C2 (×2) | 1.824(1) | 10 |
| | – Tb3 (×2) | 3.03(2) | |
| | – Tb2 (×2) | 3.05(2) | |
| | – Tb4 (×2) | 3.19(2) | |
| | – Tb4 (×2) | 3.19(2) | |
| Co2 | – C4 | 1.77(2) | 10 |
| | – C1 | 1.94(2) | |
| | – Tb3 (×2) | 2.98(2) | |
| | – Tb2 (×2) | 3.09(2) | |
| | – Tb4 (×2) | 3.10(2) | |
| | – Tb4 (×2) | 3.11(2) | |
| C1 | – Co2 | 1.94(2) | 6 |
| | – Tb1 | 2.355(9) | |
| | – Tb3 (×2) | 2.477(8) | |
| | – Tb2 (×2) | 2.530(8) | |
| C2 | – Co1 (×2) | 1.824(1) | 6 |
| | – Tb3 | 2.45(1) | |
| | – Tb4 | 2.51(1) | |
| | – Tb2 | 2.54(1) | |
| | – Tb4 | 2.57(1) | |
| C3 | – Tb1 (×4) | 2.492(6) | 6 |
| | – Tb2 (×2) | 2.600(8) | |
| C4 | – Co2 (×2) | 1.77(2) | 6 |
| | – Tb4 (×4) | 2.546(6) | |

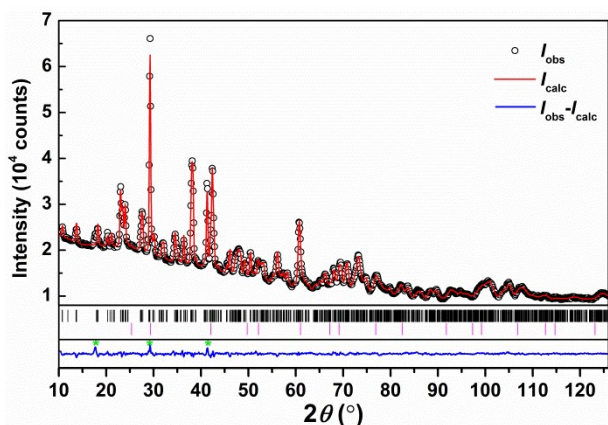**Fig. S1** Full neutron powder diffraction pattern of $Tb_4Co_2C_3$ collected at 100 K and $\lambda$ = 1.28 Å. Black ticks indicate the positions of Bragg reflections for the $Tb_4Co_2C_3$ phase, magenta ticks – $\beta$-$Tb_2C$ [structure type (ST) NaCl] phase.
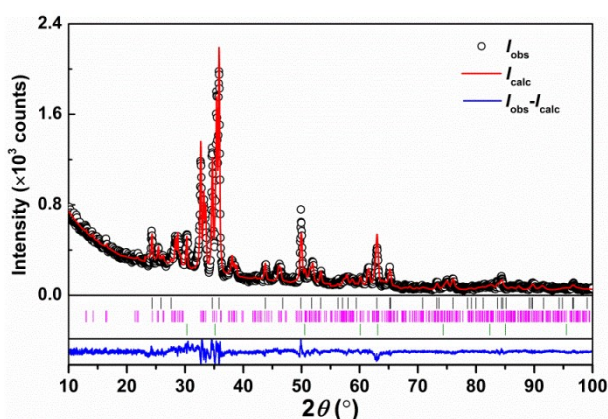


**Fig. S2** X-ray powder diffraction pattern for $Y_4Co_2C_3$ measured at 293(2) K and $\lambda$ = 1.54185 Å. Black ticks indicate the positions of Bragg reflections for the YCoC phase, magenta ticks – $Y_4Co_2C_3$, green ticks – $\beta$-$Y_2C$ (ST NaCl) phase.



**Fig. S3** X-ray powder diffraction pattern for $Gd_4Co_2C_3$ measured at 293(2) K and $\lambda$ = 1.54185 Å. Black ticks indicate the positions of Bragg reflections for the $Gd_4Co_2C_3$ phase, magenta ticks – $\beta$-$Gd_2C$ (ST NaCl) impurity phase. The green asterisk points out an impurity peak corresponding to the (311) reflection of $GdCo_2$ (ST $MgCu_2$).
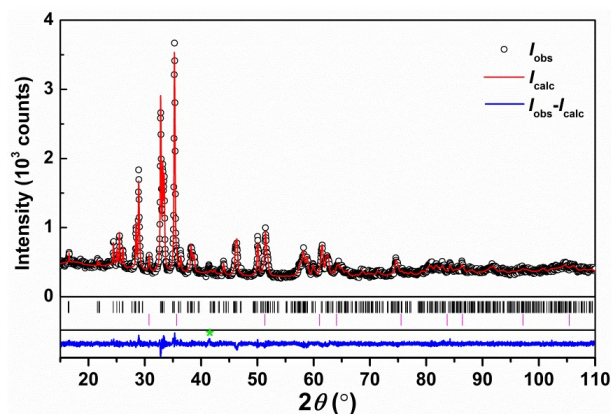


**Fig. S4** X-ray powder diffraction pattern for $Tb_4Co_2C_3$ measured at 293(2) K and $\lambda$ = 1.54185 Å. Black ticks indicate the positions of Bragg reflections for the $Tb_4Co_2C_3$ phase, magenta ticks – $\beta$-$Tb_2C$ (ST NaCl) impurity phase. The green asterisk points out an impurity peak corresponding to the (311) reflection of $TbCo_2$ (ST $MgCu_2$).
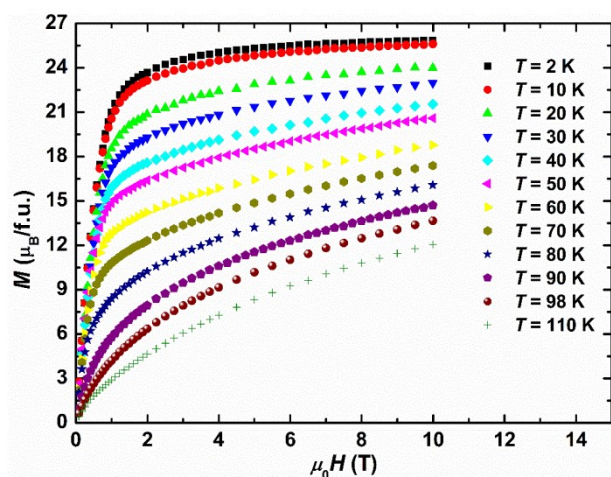


**Fig. S5** Isothermal magnetization curves of $Gd_4Co_2C_3$.



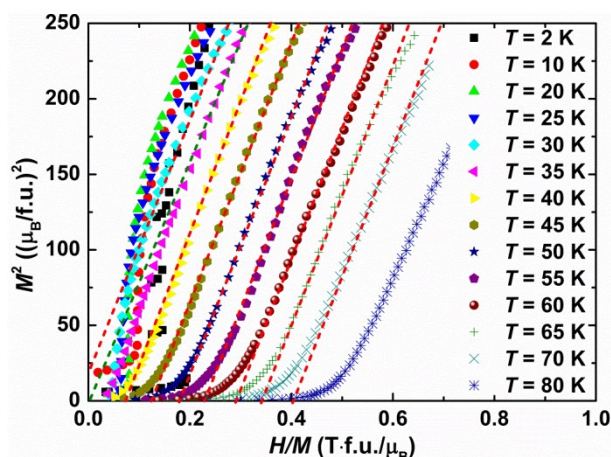**Fig. S6** Arrott-Belov plots for $Tb_4Co_2C_3$. The dashed lines are linear extrapolations.