

**Balancing structural distortions via competing 4f and itinerant interactions: A case of polymorphism in magnetocaloric  $\text{HoCo}_2$**

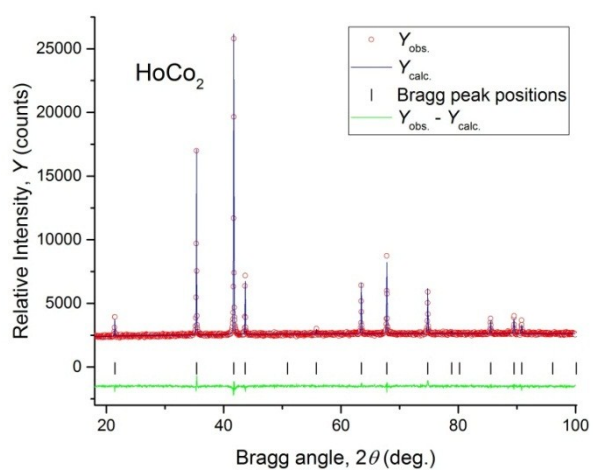


Figure S1. Room temperature X-ray Powder Diffraction Pattern of  $\text{HoCo}_2$  (II)

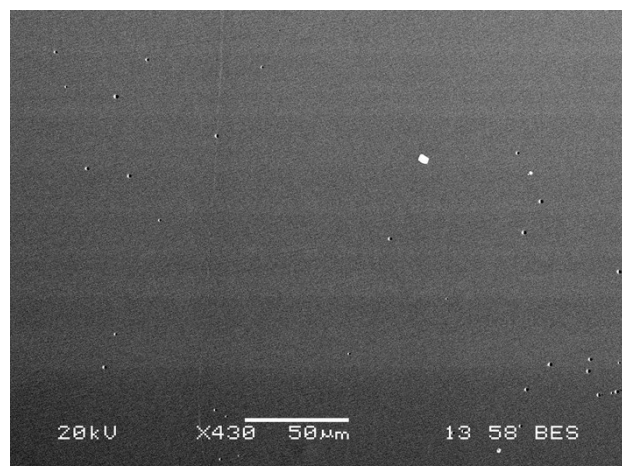


Figure S2. SEM micrograph of  $\text{HoCo}_2$  sample (II). The white spots belong to holmium oxide.

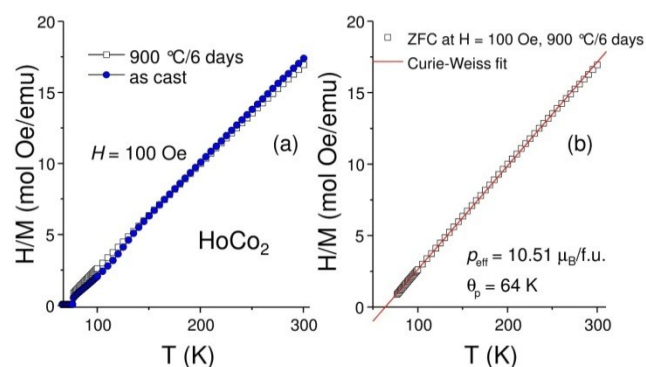


Figure S3. The inverse dc susceptibility of  $\text{HoCo}_2$ : a) a comparison of as cast and heat-treated samples; b) Curie-Weiss fit of the heat-treated sample

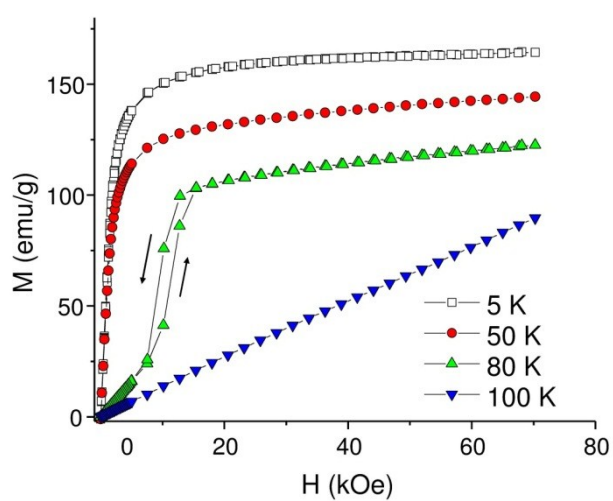


Figure S4. The Magnetization as a function of applied magnetic field measured for  $\text{HoCo}_2$  at 5, 50, 80, and 100 K.

Table S1. Crystal structures observed in  $\text{HoCo}_2$ .

T, K	Space Group	$a$ , Å	$b$ , Å	$c$ , Å	Ho: $x,y,z$	Co: $x,y,z$
300	$\overline{\text{Fd}}\ 3\text{m}$	7.1786(1)	7.1786(1)	7.1786(1)	5/8, 5/8, 5/8	0, 0, 0
50	$I4_1/amd$	5.0718(1)	5.0718(1)	7.1552(2)	0, 1/4, 3/8	0, 0, 0
10	Fddd	7.1742(2)	7.163(1)	7.161(1)	5/8, 5/8, 5/8	0, 0, 0
10	$\text{Imma}^*$	5.0610(1)	5.0671(1)	7.1753(2)	0, 1/4, 0.376	Co1:0,0,0 Co2:1/4,3/4,1/4

\*Refined as potential crystal structure; used for the DFT calculations

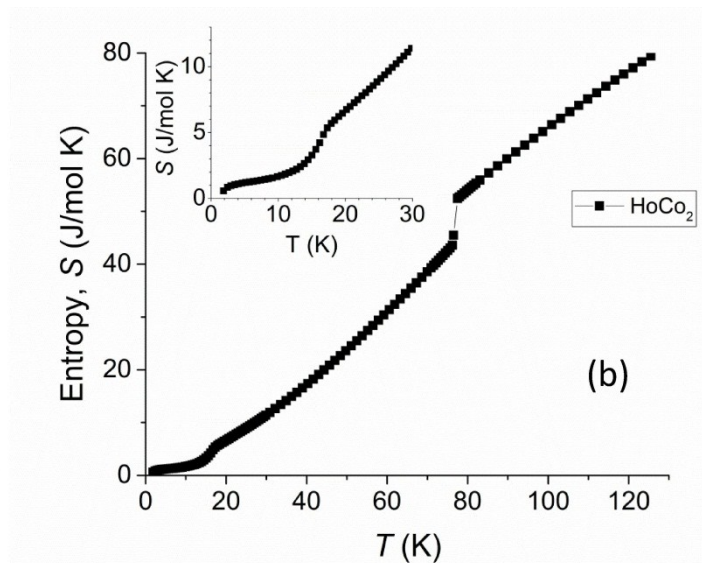
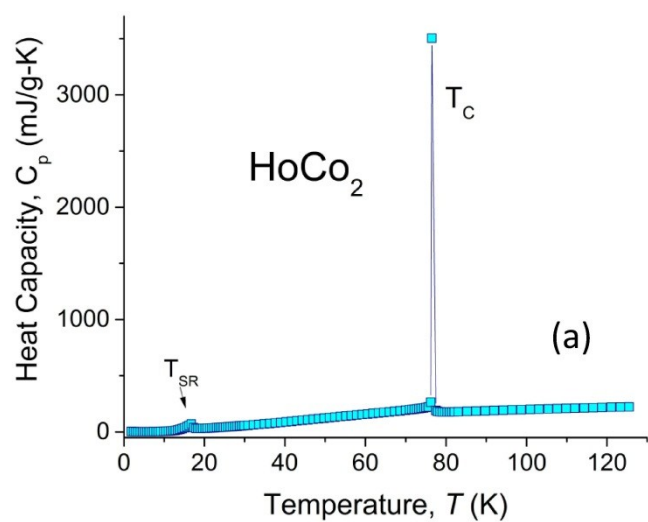


Figure S5. (a) Heat capacity and (b) Entropy of  $\text{HoCo}_2$  as a function of temperature between 2 and 125 K in zero magnetic field. Note: Entropy plot has a small systematic error due to not accounting for the heat capacity of  $\text{HoCo}_2$  between 0 and 2 K but it can be used to verify the order of the transitions.