Supplementary Information

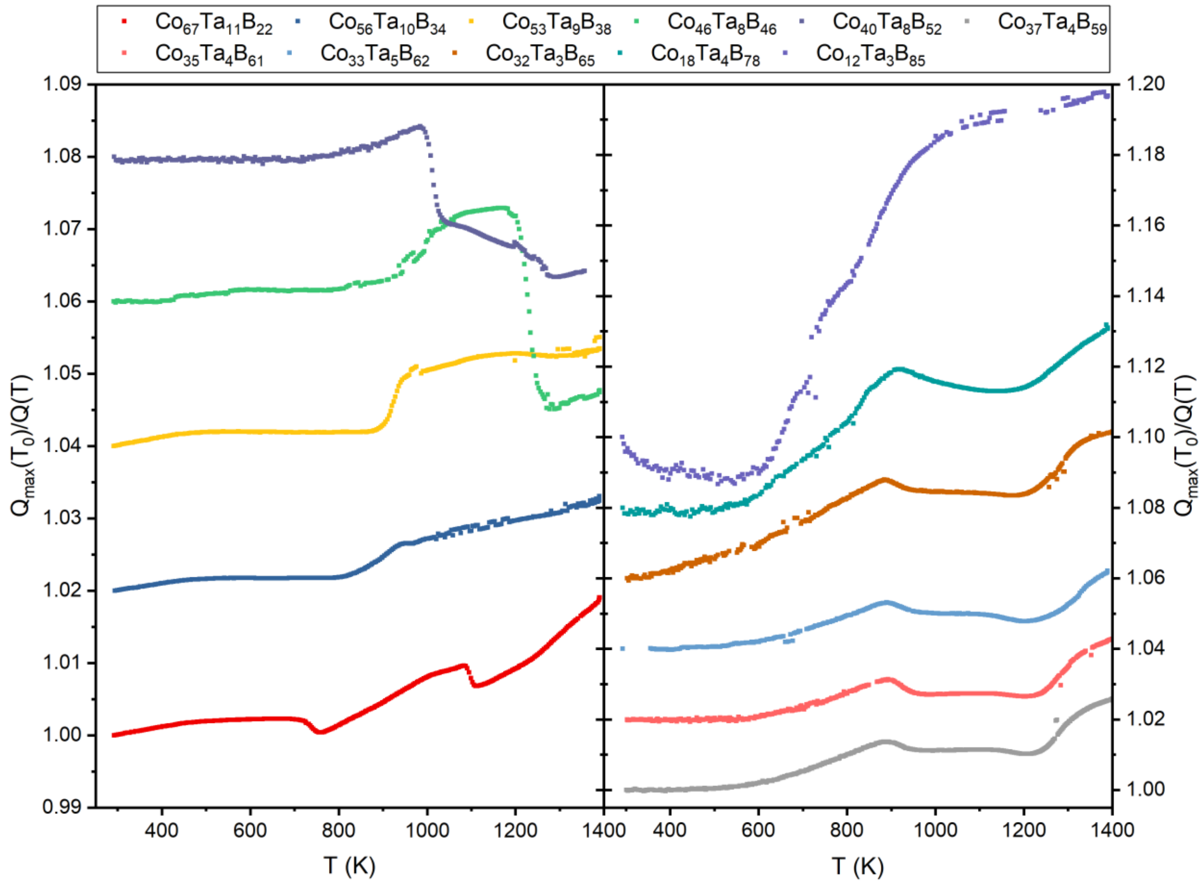

Figure S1. Shift of the principal peak of the structure factor with temperature during *in situ* heating dependent on the chemical composition of $(Co_{6.8}Ta)_{100-x}B_x$. The curves are offset to enhance the clarity of the figure.

The glass transition temperature was determined by the "change-in-slope" method based on the fact that at T_g, free volume begins to increase [1,2]. Since the shift of the principle peak of the structure factor, $Q(T_0)/Q(T)$, is proportional to the volume change $(V/V_0)^{1/3}$ [1], a change to a (more) positive slope of $Q(T_0)/Q(T)$ is expected at T_g.

For B concentrations larger 46 at.%, the $Q(T_0)/Q(T)$ curves (Fig. S1) reveal a smaller change in slope at T_g. This different character of the glass transition, together with different T_g and S_c hints to structural differences between CoTaB with high (>59 at.%) and low (<46 at.%) B concentration, while the region in between is proposed to be a transition region between both regimes.

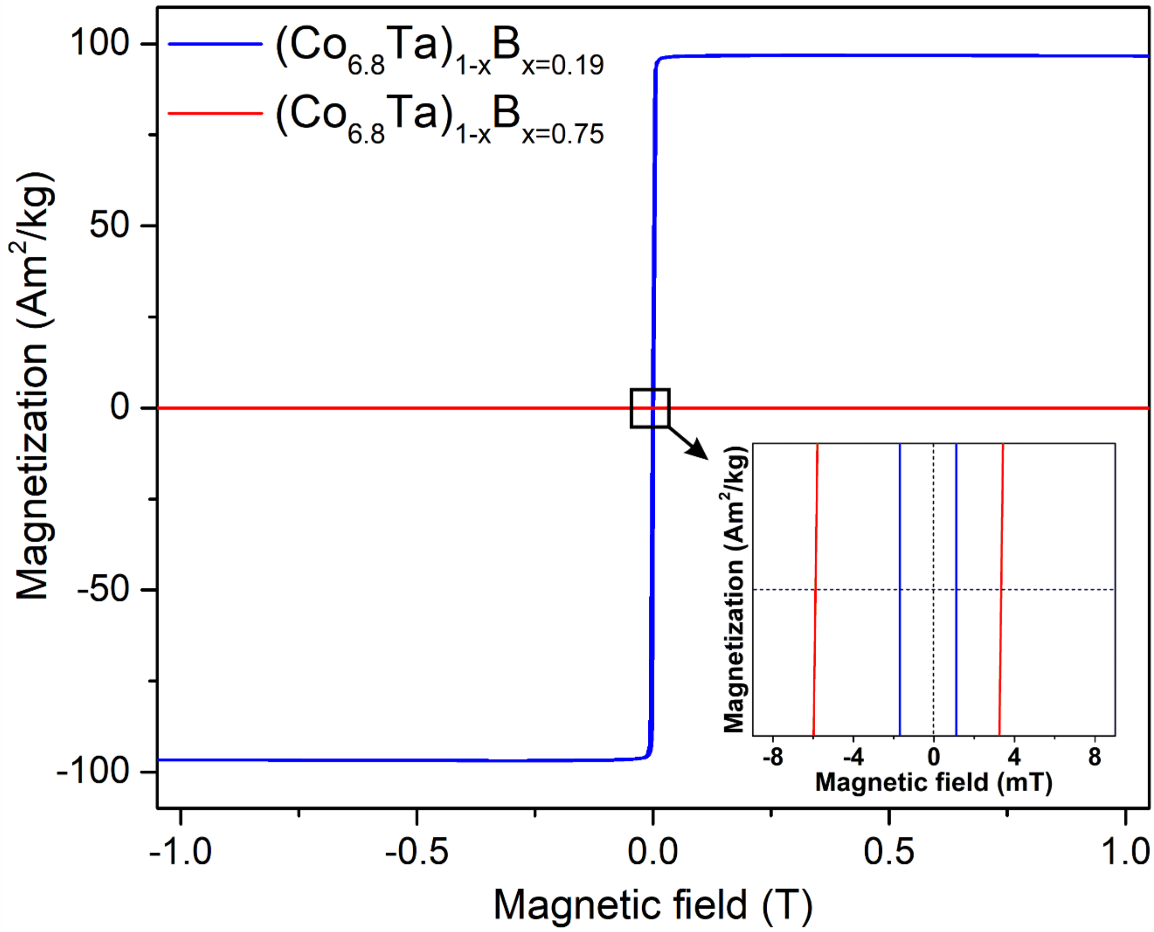

Figure S2. Hysteresis curve of $Co_{67}Ta_{14}B_{19}$ and $Co_{21}Ta_{4}B_{75}$ measured at room temperature in a vibrating sample magnetometer.

References

1. Yavari, A.R.; Le Moulec, A.; Inoue, A.; Nishiyama, N.; Lupu, N.; Matsubara, E.; Botta, W.J.; Vaughan, G.; Di Michiel, M.; Kvick, Å. Excess free volume in metallic glasses measured by X-ray diffraction. *Acta Mater.* **2005**, *53*, 1611–1619, doi:10.1016/j.actamat.2004.12.011.
2. Turnbull, D.; Cohen, M.H. Free-Volume Model of the Amorphous Phase: Glass Transition. *J. Chem. Phys.* **1961**, *34*, 120–125.