supplementary information

A case of multifunctional intermetallic compounds: negative thermal expansion coupling with magnetocaloric effect in $(Gd,Ho)(Co,Fe)_2$

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Supplementary Figures

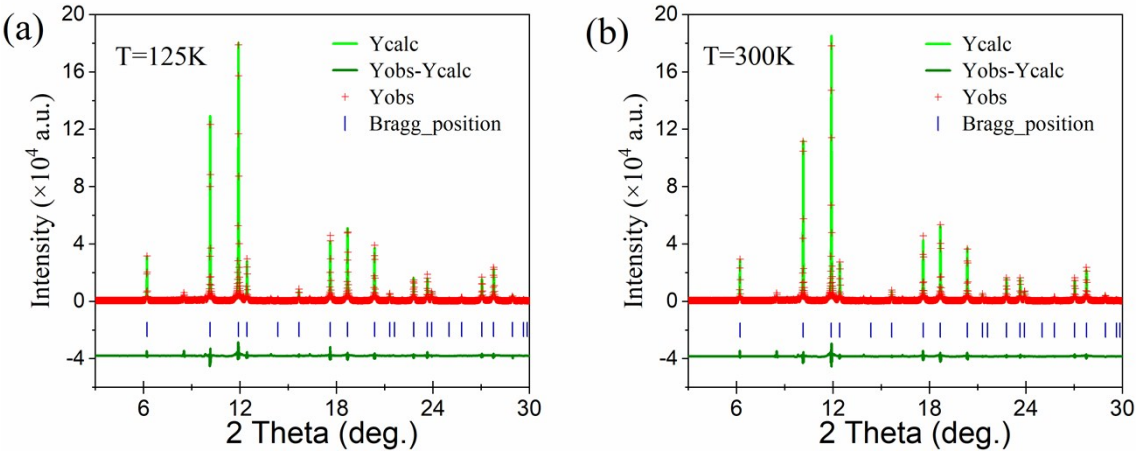

Figure S1. (a, b) Rietveld refinement patterns of high-resolution synchrotron X-ray powder diffraction (SPD) data for $Gd_{0.3}Ho_{0.7}Co_{1.92}Fe_{0.08}$ (GHCF) at 125 K and 300 K: the patterns were fitted using the cubic structure (RIKEN SPring-8 Center, Japan; λ= 0.45 Å). There is no peak split in the NPD patterns for GHCF from 10 to 350 K (**Figure** 1b), but minor split was observed in the high-resolution synchrotron X-ray powder diffraction patterns (SPD),[1] suggesting GHCF is a pseudo-cubic phase below T_C and changes to cubic above T_C.

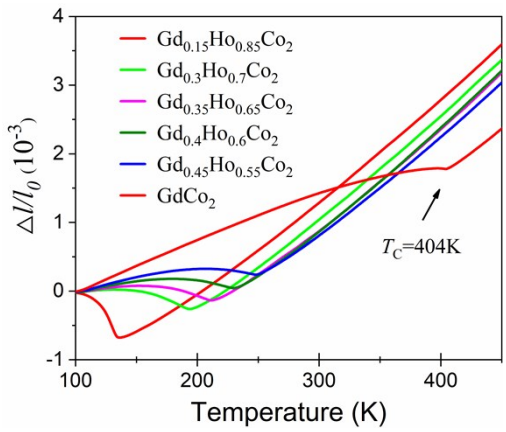

Figure S2. Thermal expansion of $Gd_xHo_{1-x}Co_2$ determined by a thermo-dilatometer. The linear thermal expansion curves ($\Delta l/l_0$) were measured by an advanced dilatometer (NETZSCH DIL402) with heating rate of 5 K/min.

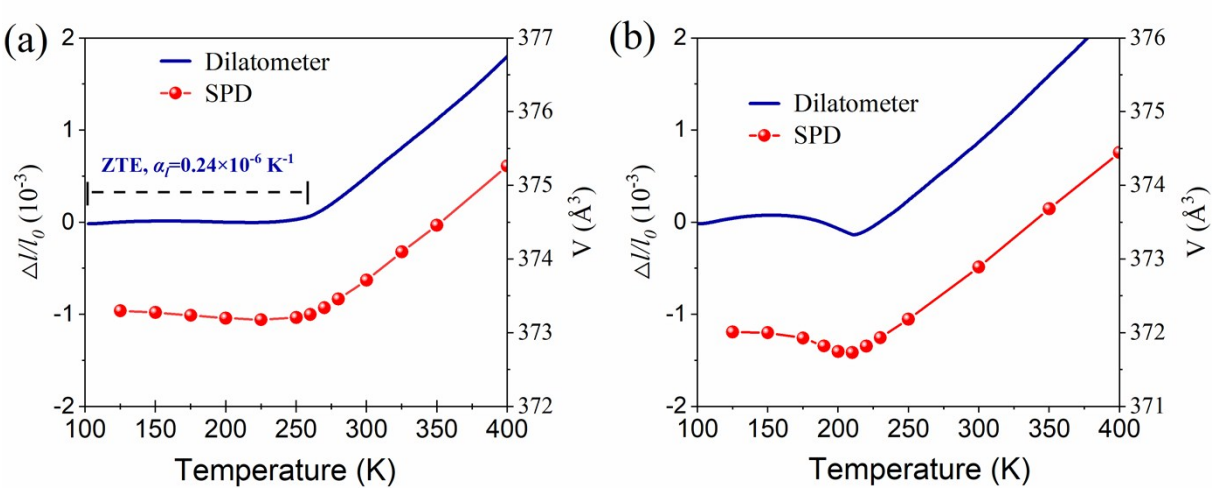

Figure S3. Thermal expansion of (a) $Gd_{0.3}Ho_{0.7}Co_{1.92}Fe_{0.08}$ (GHCF) and (b) $Gd_{0.35}Ho_{0.75}Co_2$ confirmed by both dilatometer and high-resolution synchrotron X-ray powder diffraction experiments.

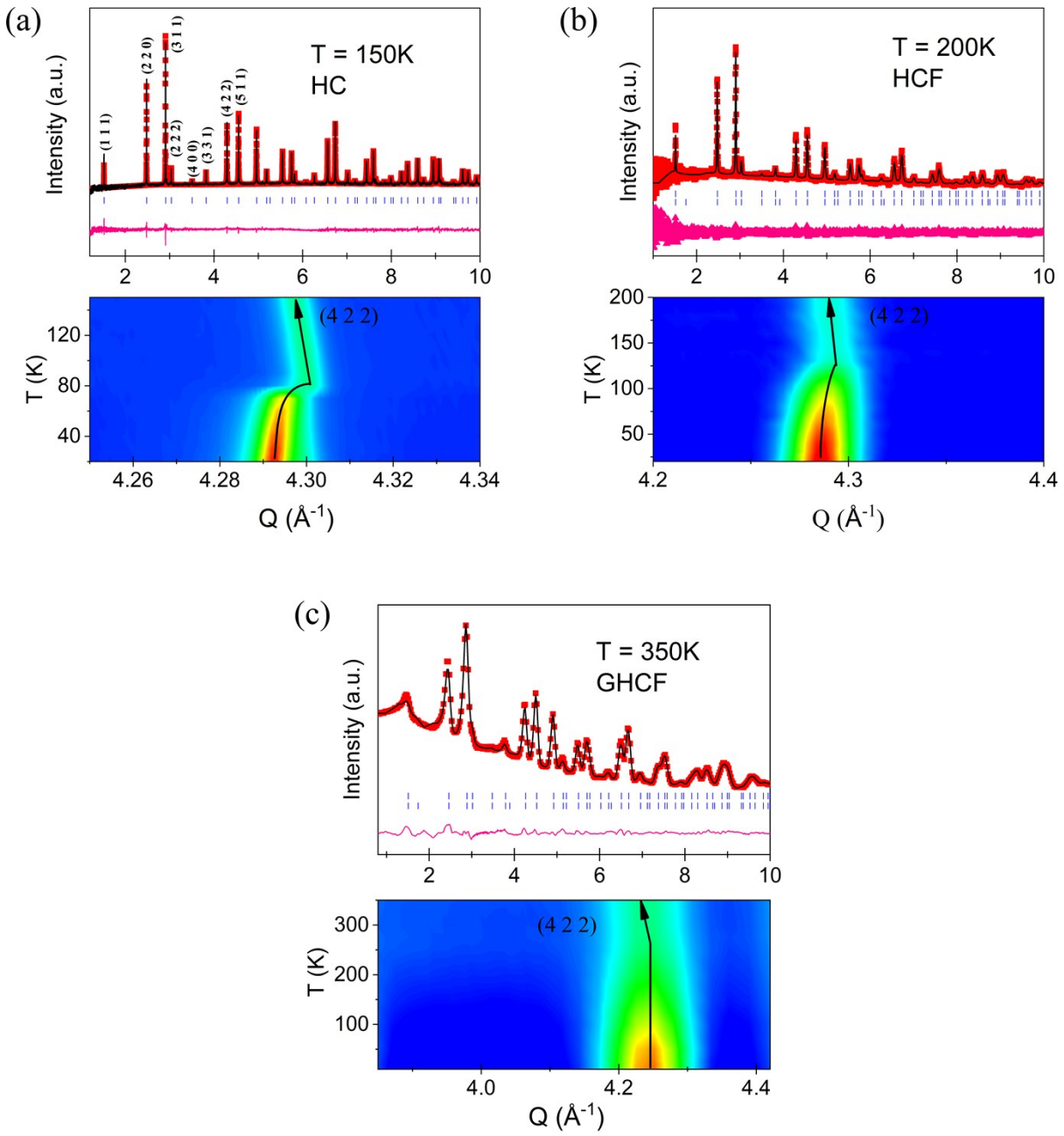

Figure S4. Structure refinements of NPD patterns and contour plots of the (4 2 2) profile intensity for (a) $HoCo_2$ (HC), (b) $HoCo_{1.92}Fe_{0.08}$ (HCF) and (c) $Gd_{0.3}Ho_{0.7}Co_{1.92}Fe_{0.08}$ (GHCF).

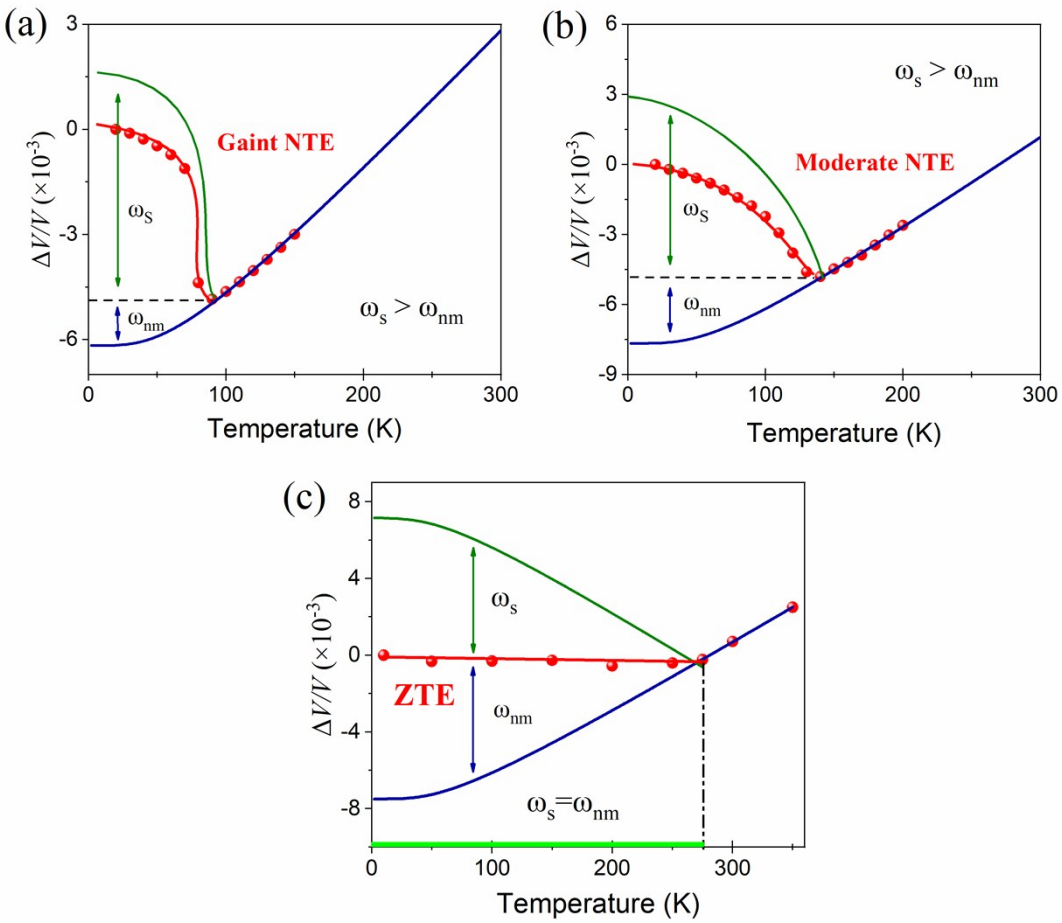

Figure S5. Experimental and calculated thermal expansion of (a) HC, (b) HCF, and (c) GHCF. The ω_{exp} is the relative ratio of experimental volume, and ω_{nm} is nominal thermal expansion on the basis of Debye-Grüneisen model.

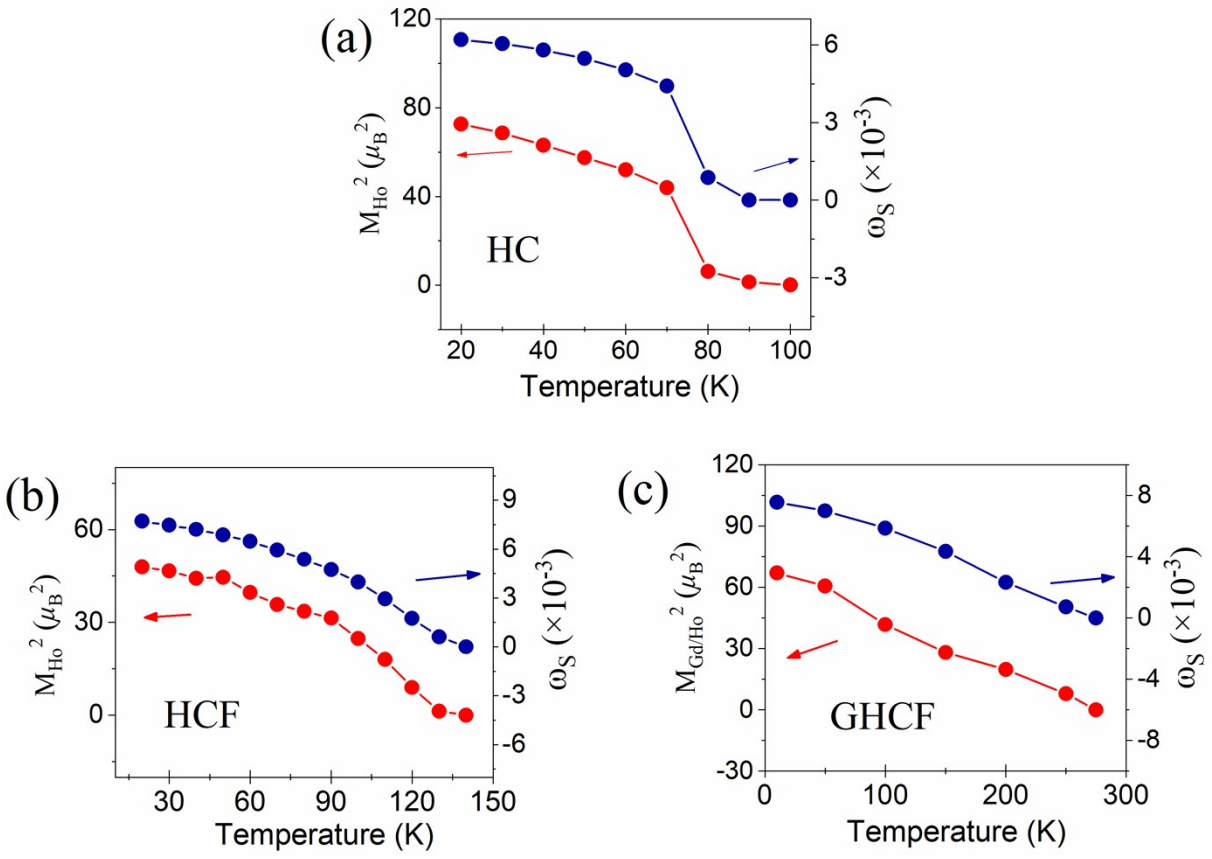

Figure S6. Quantitative relationship between M_R^2 and ω_s in (a) HC, (b) HCF and (c) GHCF

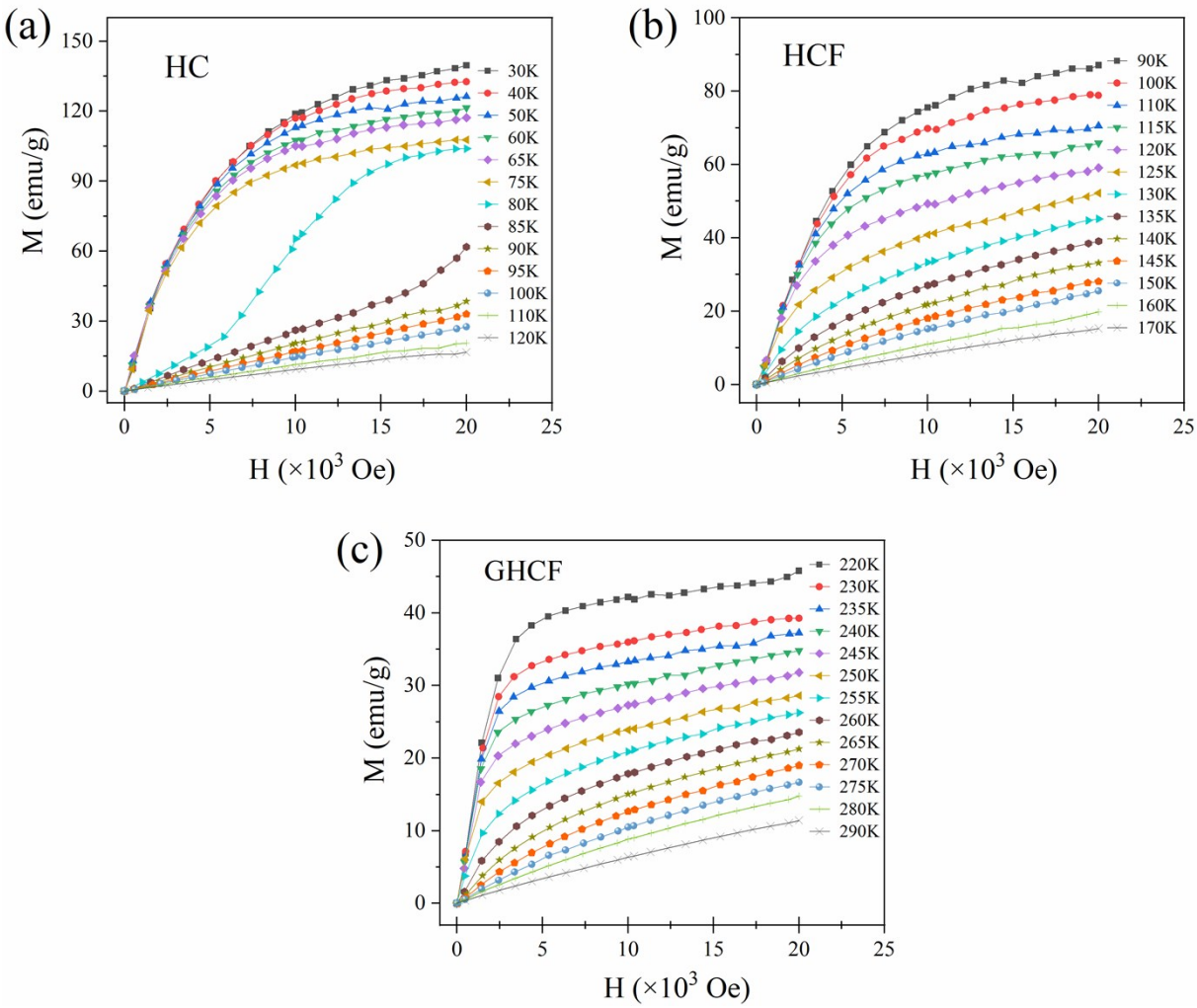

Figure S7. Isothermal *M-H* curves (0 - 20 KOe) for (a) HC, (b) HCF and (c) GHCF.

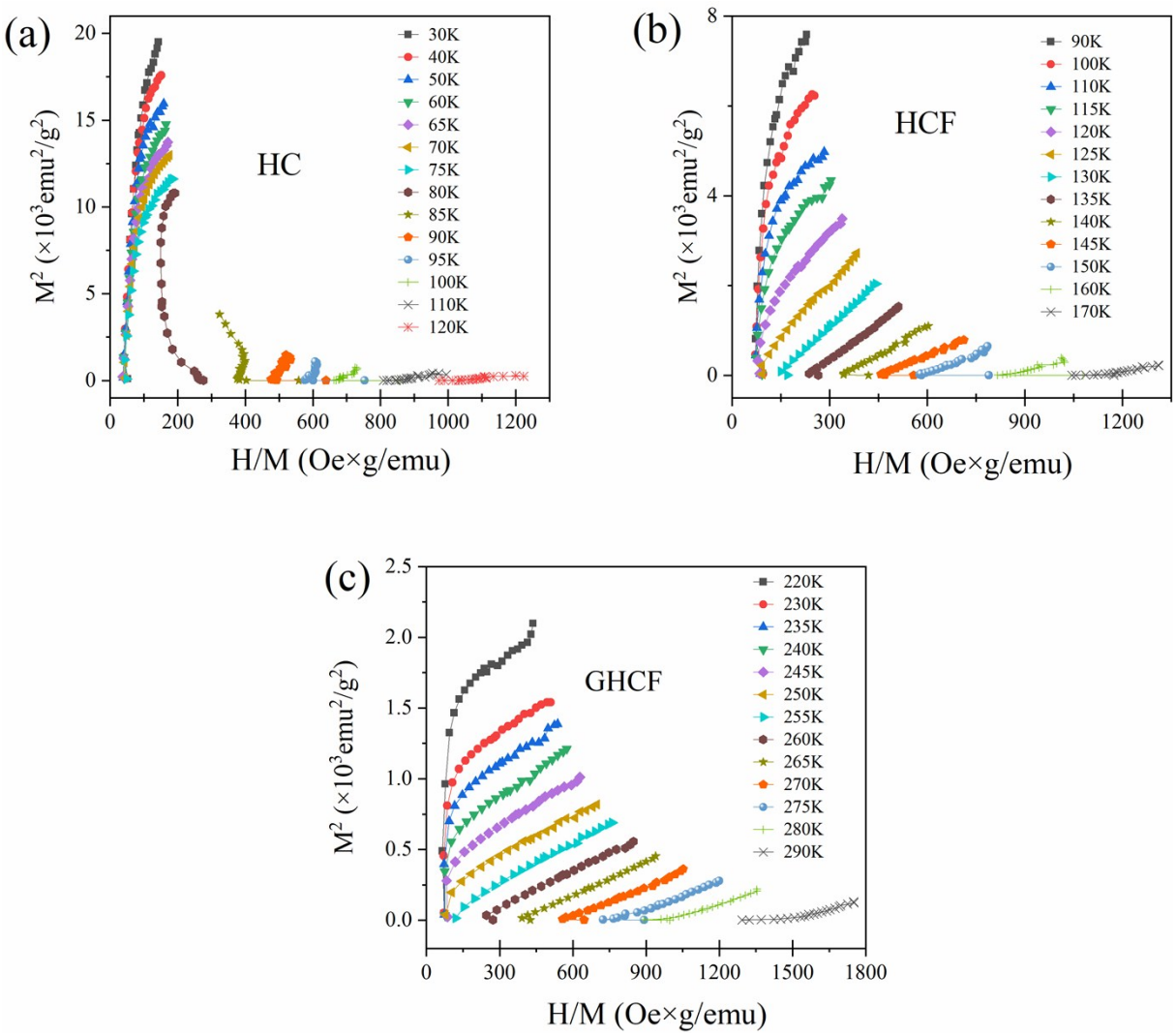

Figure S8. Isothermal Arrott plots of M^2 versus H/M derived from M-H curves for (a) HC, (b) HCF and (c) GHCF.

Reference

1 K. Kato, Y. Tanaka, M. Yamauchi, K. Ohara and T. Hatsui, *J. Synchrotron Radiat.*, 2019, **26**, 762-773.