

## Supporting Information

### Composition-Dependent Magnetic Anisotropy in Cu/Mn-Based Two-Dimensional Hybrid Perovskite Single Crystals

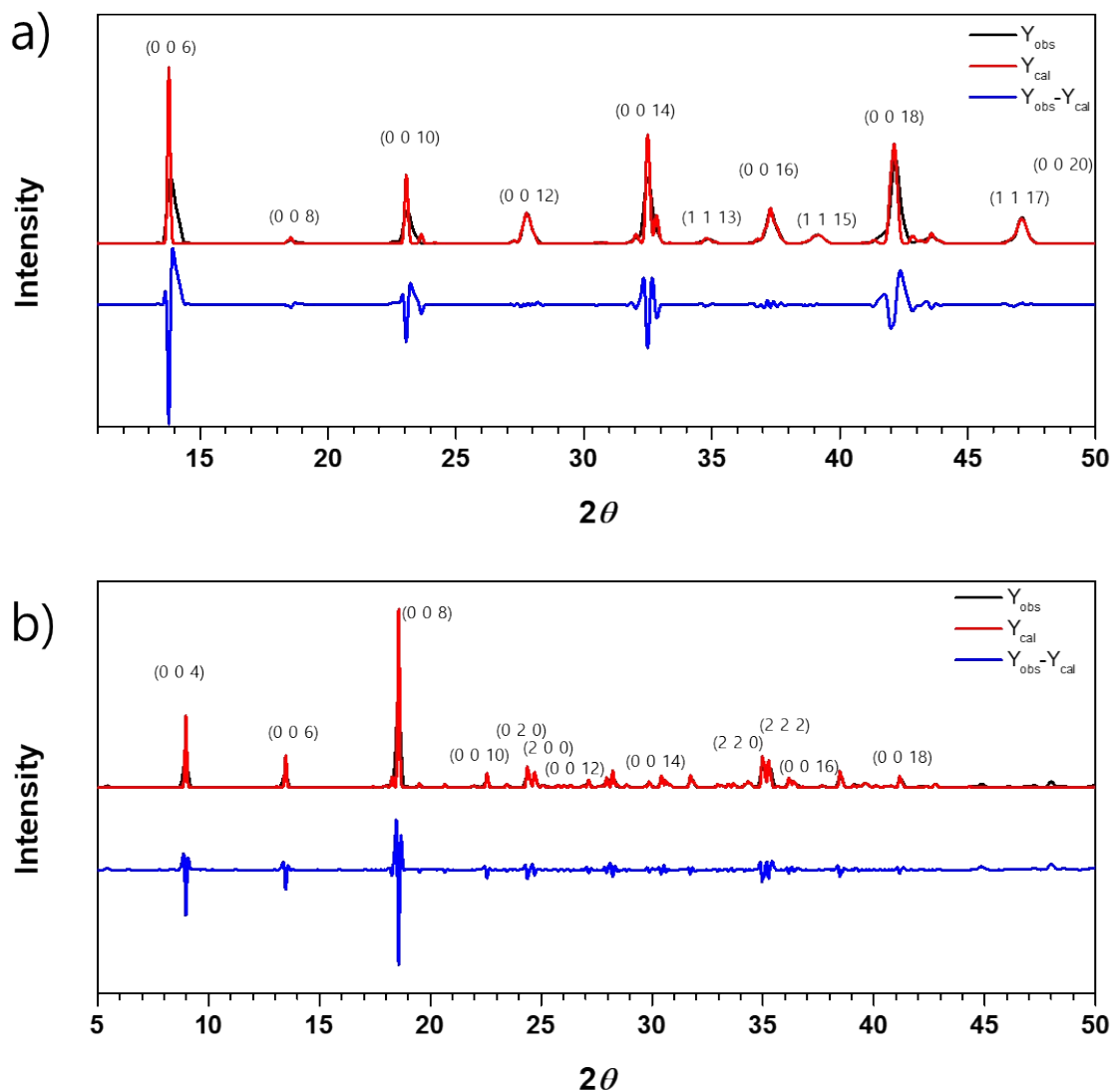
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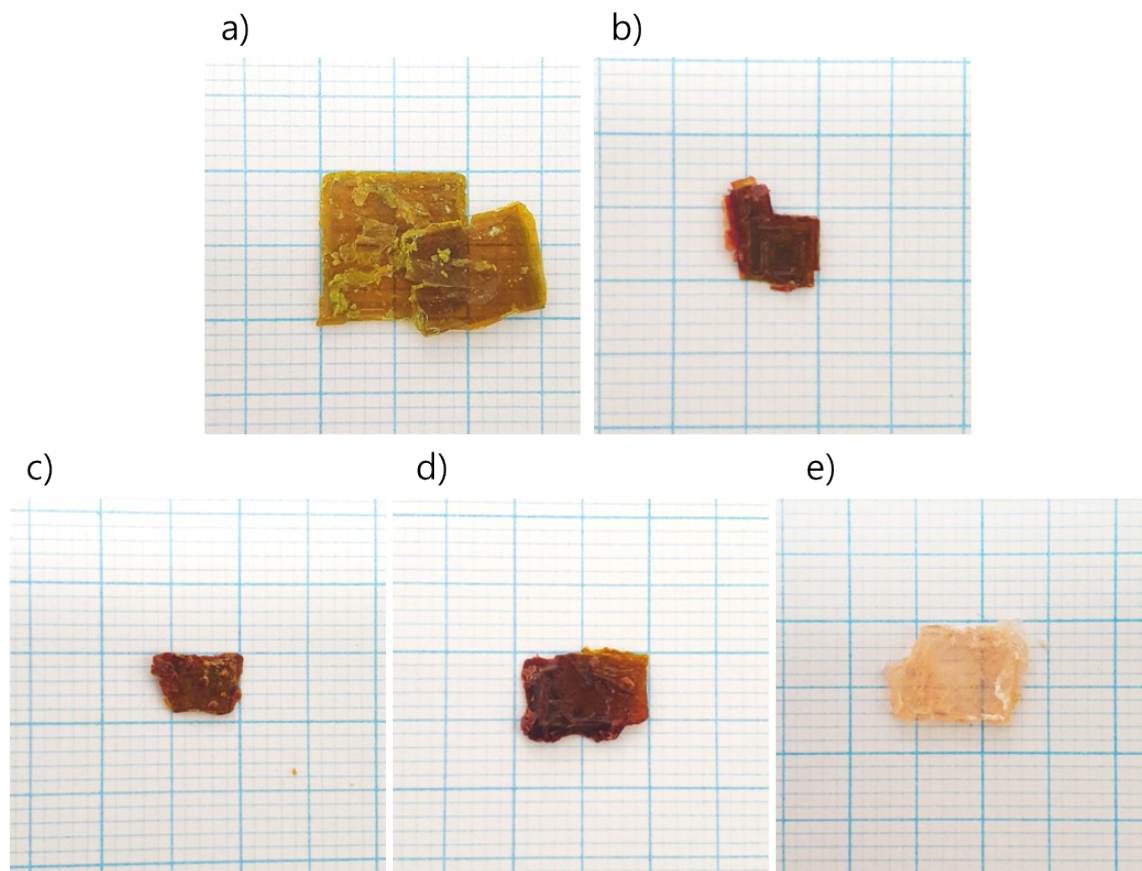
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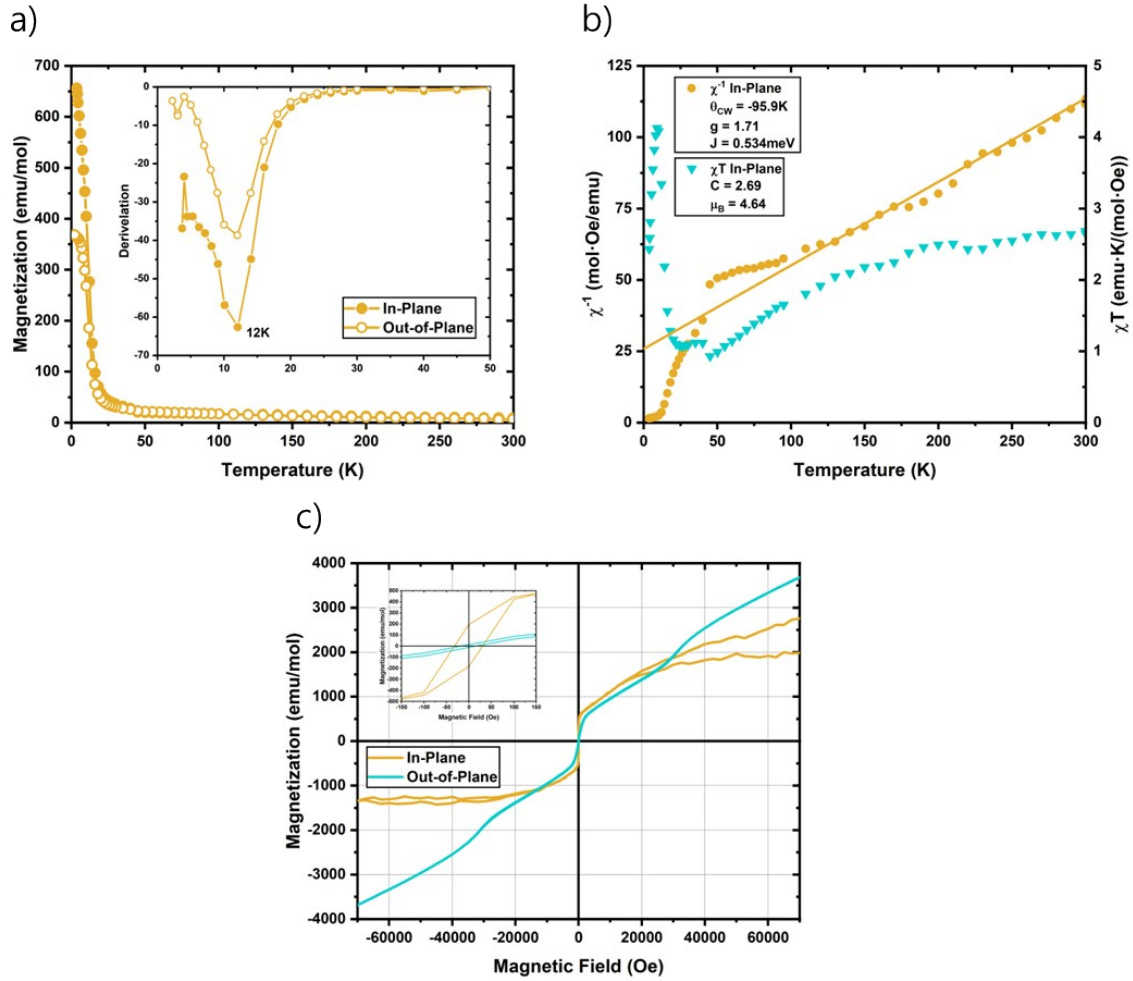
**Figure S1.** Rietveld refinement profiles of (a) Cu-PEA and (b) Mn-PEA single crystals measured at room temperature. The observed ( $Y_{obs}$ , black), calculated ( $Y_{cal}$ , red), and difference ( $Y_{obs} - Y_{cal}$ , blue) patterns are shown. The indexed (h k l) reflections confirm the orthorhombic symmetry ( $Pbca$ ) and support the reliable determination of lattice parameters. The elevated R-factors ( $R_p=37.6\%$  for Cu-PEA and  $R_p=40.2\%$  for Mn-PEA) are attributed to strong preferred orientation arising from the plate-like single crystal morphology, which leads to intensity mismatch predominantly along non-(001) reflections. The  $2\theta$  ranges between panels to capture the relevant diffraction features of each compound.

Sample	$(h\ k\ l)$	$2\theta(^{\circ})$	$FWHM(^{\circ})$	Crystallite Size (nm)
Cu-PEA	(0 0 4)	9.168	0.231	34.4
Mn-PEA	(0 0 4)	8.961	0.225	35.2

**Table S1.** Crystallite sizes of Cu-PEA and Mn-PEA. The values were calculated using the Scherrer equation based on the  $2\theta$  positions and FWHM of the low-angle (0 0 4) reflection peaks.



**Figure S2.** Optical images of single crystals of a) Cu-PEA, b)  $\text{Cu}_{0.75}\text{Mn}_{0.25}$ -PEA, c)  $\text{Cu}_{0.50}\text{Mn}_{0.50}$ -PEA, d)  $\text{Cu}_{0.25}\text{Mn}_{0.75}$ -PEA, and e) Mn-PEA. All samples crystallize as square plate-like single crystals, consistent with the layered nature of the Ruddlesden–Popper structure. A gradual change in crystal coloration is observed with increasing Mn content, reflecting compositional variation across the series.



**Figure S3.** Magnetic characterization of  $\text{Cu}_{0.17}\text{Mn}_{0.83}\text{-PEA}$  single crystals.

a) Temperature-dependent magnetization ( $M$ - $T$ ) measured in both in-plane and out-of-plane configurations under an applied field of 1000 Oe. A low-temperature magnetic transition is observed near 12 K.

b) Temperature dependence of inverse susceptibility ( $\chi^{-1}$ ) and  $\chi T$  with fitting to the Curie-Weiss law and two-dimensional Heisenberg model. The analysis yields a negative Weiss temperature, indicating dominant antiferromagnetic interactions, while finite exchange parameters reflect competing magnetic exchange contributions within the layered framework.

c) Field-dependent magnetization ( $M$ - $H$ ) curve at 2 K. The data show an initial hysteretic response followed by a field-induced increase in magnetization consistent with a spin-reorientation (spin-flop-like) transition.