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Supplementary Information for

Dependence of Phase Transitions on Halide Ratio in Inorganic $CsPb(Br_xI_{1-x})_3$ Perovskite Thin Films Obtained from High-Throughput Experimentation

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Combinatorial printing

In order to construct the images with varying coverage, an algorithm was developed which constructs base matrices of certain coverages that can then be combined to the required image. The algorithm starts with an empty base, i.e. a matrix of zeros, with side length, l, such that the levels of coverage, d, is the side squared plus one, $d=l^2+1$, and then iteratively adds a one to the matrix such that the distance to the closest one is maximized. After half the matrix is full the rest of the coverages are achieved by inverting (switching zeros and ones) the lower coverage matrices. The two images for the investigated sample were constructed using base images of side length four out of which every other base was used, resulting in nine levels of coverage from full to none.

GIWAXS measurement

The chamber was placed on an x, y, z-stage in a goniometer and cooled with 18 ˚C water and flushed with room temperature nitrogen gas at 0.2 L/min. A K-type thermocouple was placed on top of the sample to record the temperature, a Dectris Pilatus 1M 2D detector was placed behind the sample chamber and a Si PIN diode above it. The sample was irradiated by X-rays from a metaljet source with a Ga:In (75:25) alloy, the diffraction was detected by the Pilatus whilst the PIN diode detected the fluorescence. To remove the Ga Kβ radiation a 25 μm Zn foil was placed before the 900 mm focal distance polycapillary which focused the X-rays through a vacuum flight tube on to the sample resulting in a horizontal spot size of ~1 mm. The x, y, z-stage was cycled through the nine different compositions, as well as one position measuring the quartz substrate, with a hold time of 10 s. The thermocouple sample temperature was calibrated by an ex-situ measurement, under the same conditions, using a thermal camera and a ZnSe window.

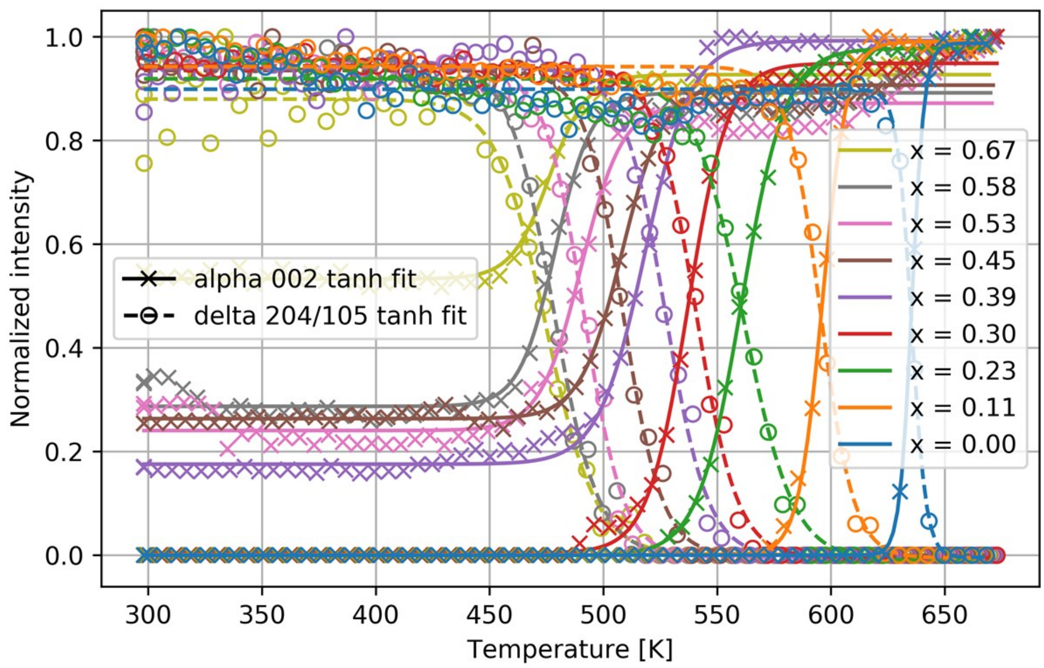

Fig. S1 The normalized integrated intensity of the 002 peak of the α-phase (crosses) and 204/105 peaks of the δ-phase (circles) as well as the sigmoid fit as a function of heating temperature.

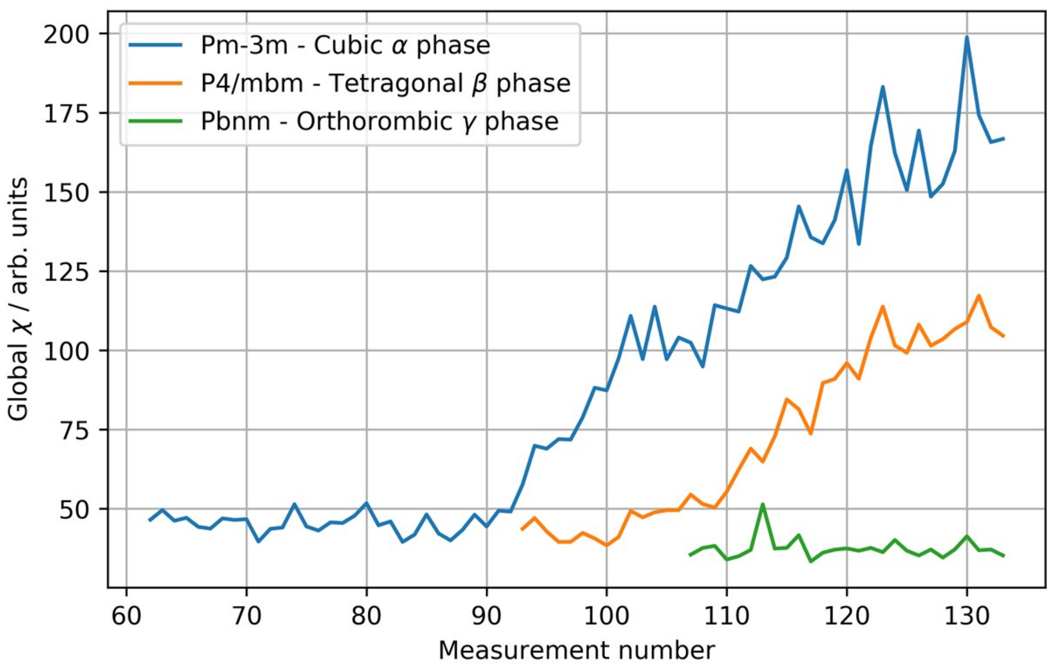

Fig. S2 The global χ of the sequential Fullprof refinement for the x = 0.23 sample.

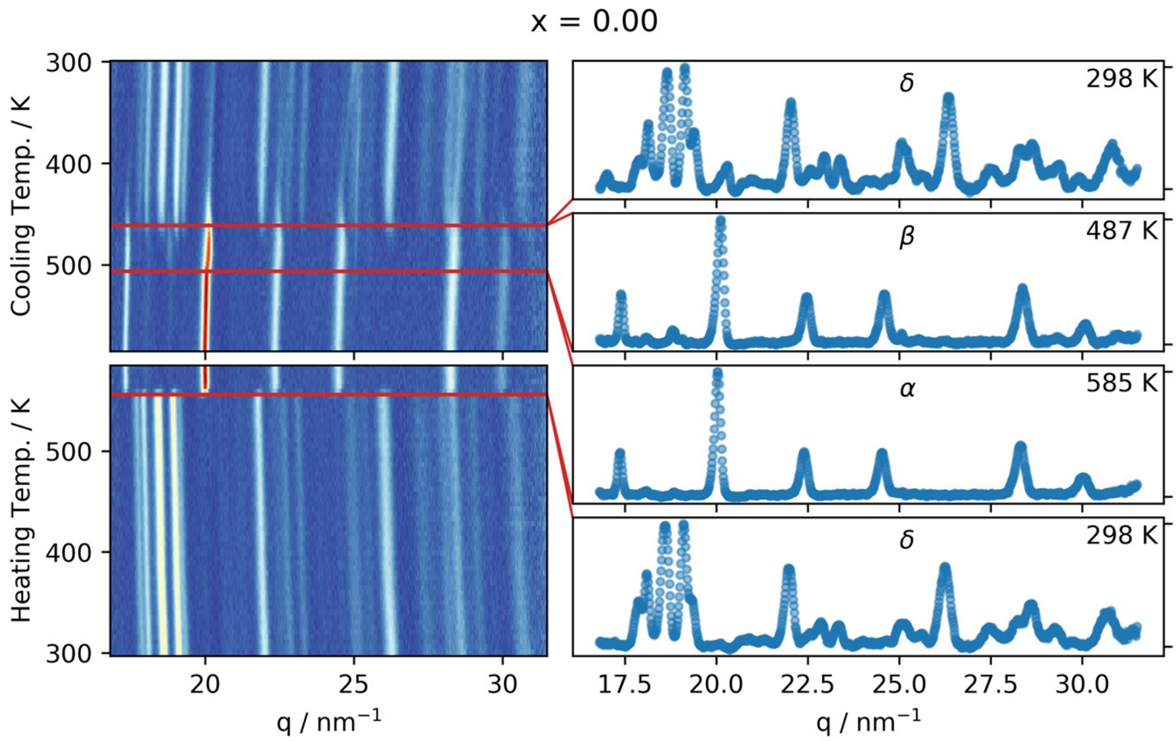

Fig. S3 Left, integrated GIWAXS pattern as a function of temperature for $CsPb(Br_xI_{1-x})_3$ with $x = 0.00$ (determined by in-situ XRF) as the sample is heated and then cooled at a rate of 2.5 K/min. Red lines indicate the phase transitions. Right, integrated GIWAXS patterns for the four different phases in the sample.

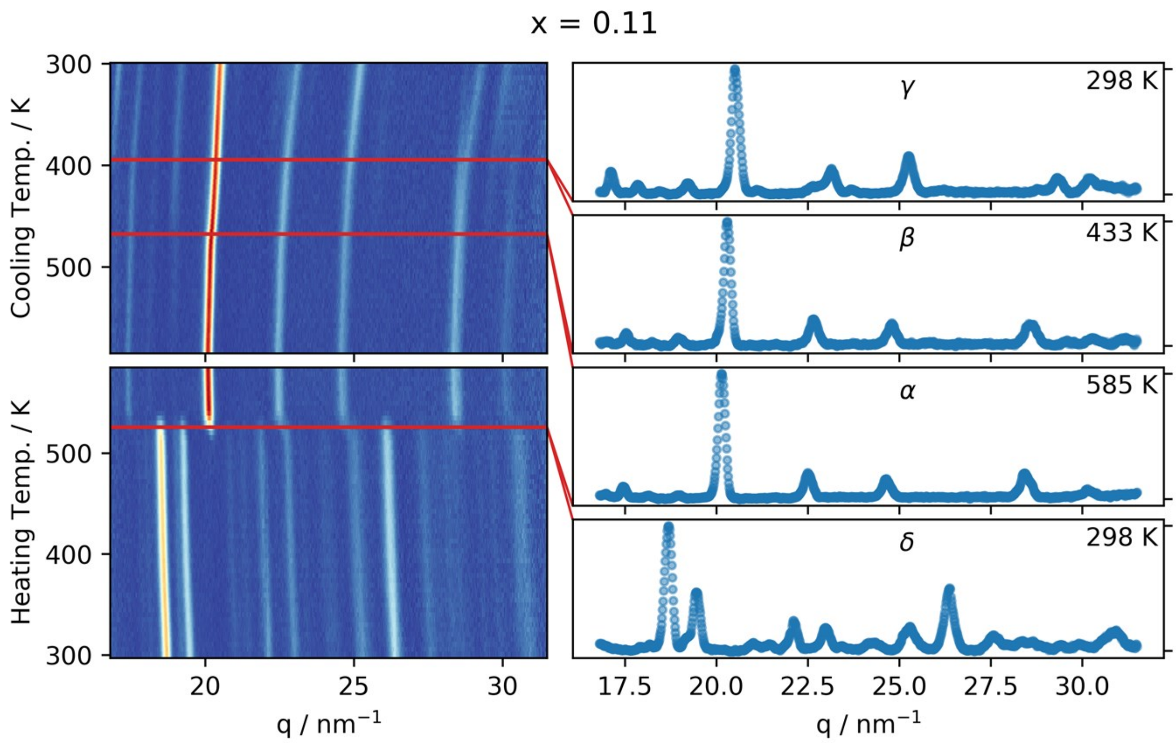

Fig. S4 Left, integrated GIWAXS pattern as a function of temperature for $CsPb(Br_xI_{1-x})_3$ with $x = 0.11$ (determined by in-situ XRF) as the sample is heated and then cooled at a rate of 2.5 K/min. Red lines indicate the phase transitions. Right, integrated GIWAXS patterns for the four different phases in the sample.

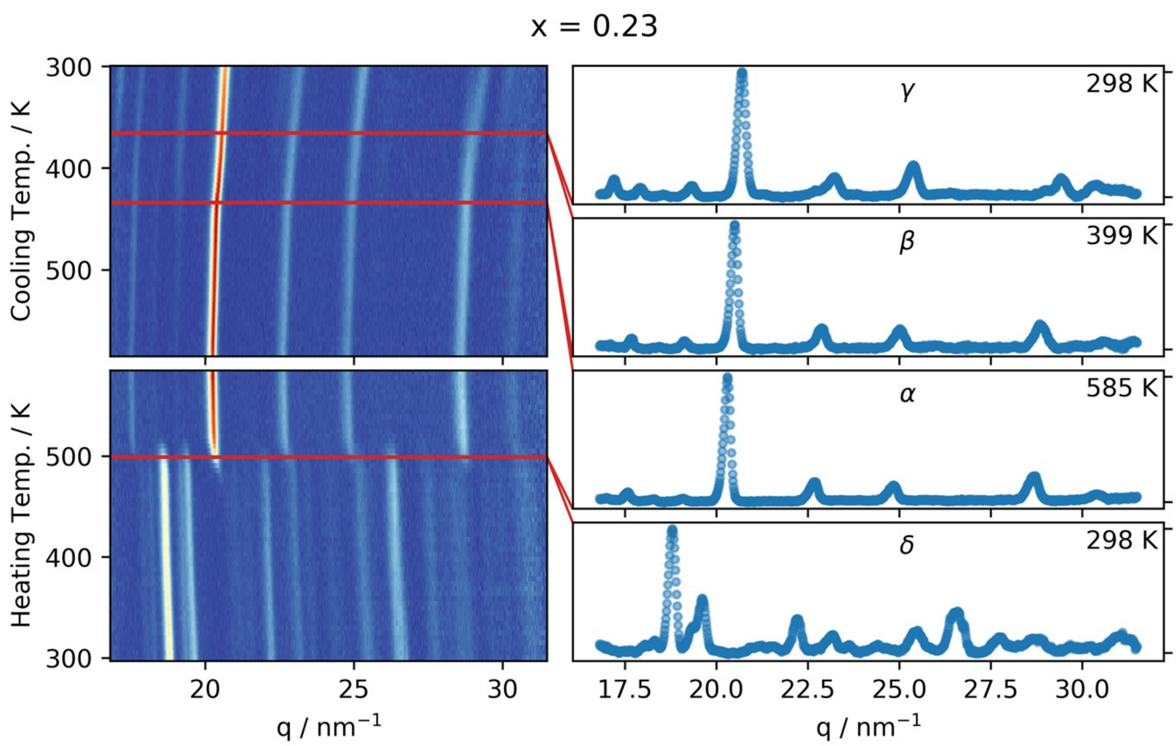

Fig. S5 Left, integrated GIWAXS pattern as a function of temperature for $CsPb(Br_xI_{1-x})_3$ with $x = 0.23$ (determined by in-situ XRF) as the sample is heated and then cooled at a rate of 2.5 K/min. Red lines indicate the phase transitions. Right, integrated GIWAXS patterns for the four different phases in the sample.

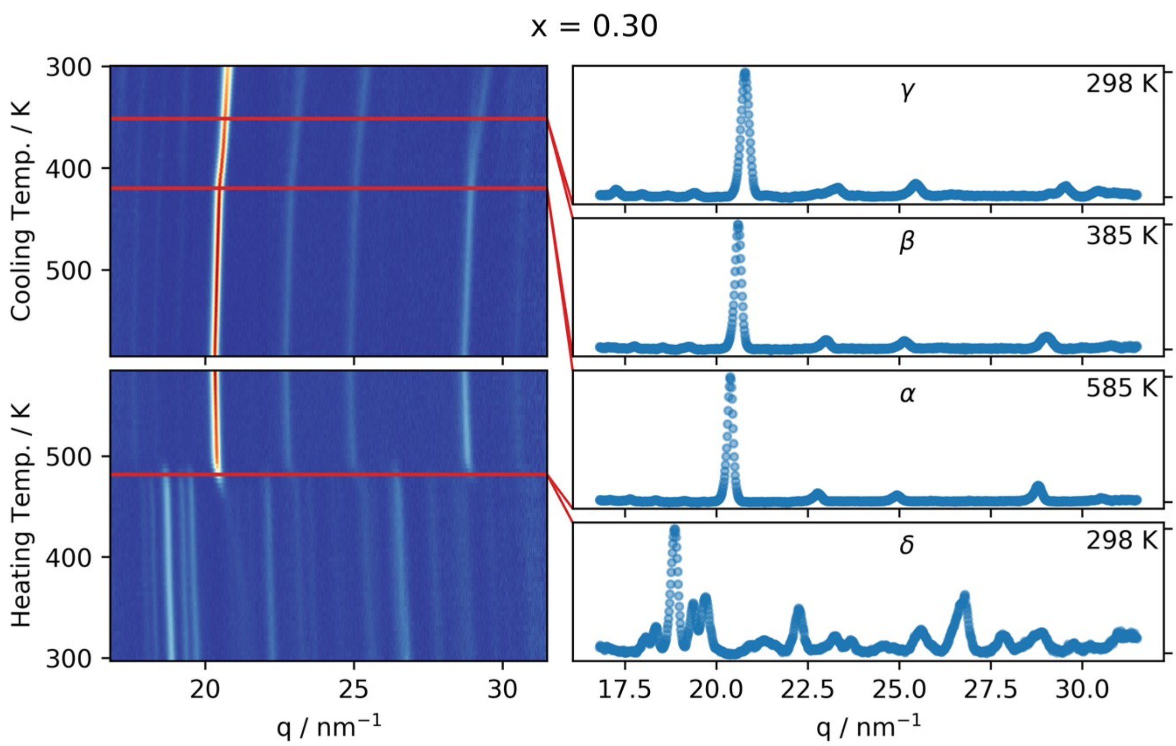

Fig. S6 Left, integrated GIWAXS pattern as a function of temperature for $CsPb(Br_xI_{1-x})_3$ with $x = 0.30$ (determined by in-situ XRF) as the sample is heated and then cooled at a rate of 2.5 K/min. Red lines indicate the phase transitions. Right, integrated GIWAXS patterns for the four different phases in the sample.

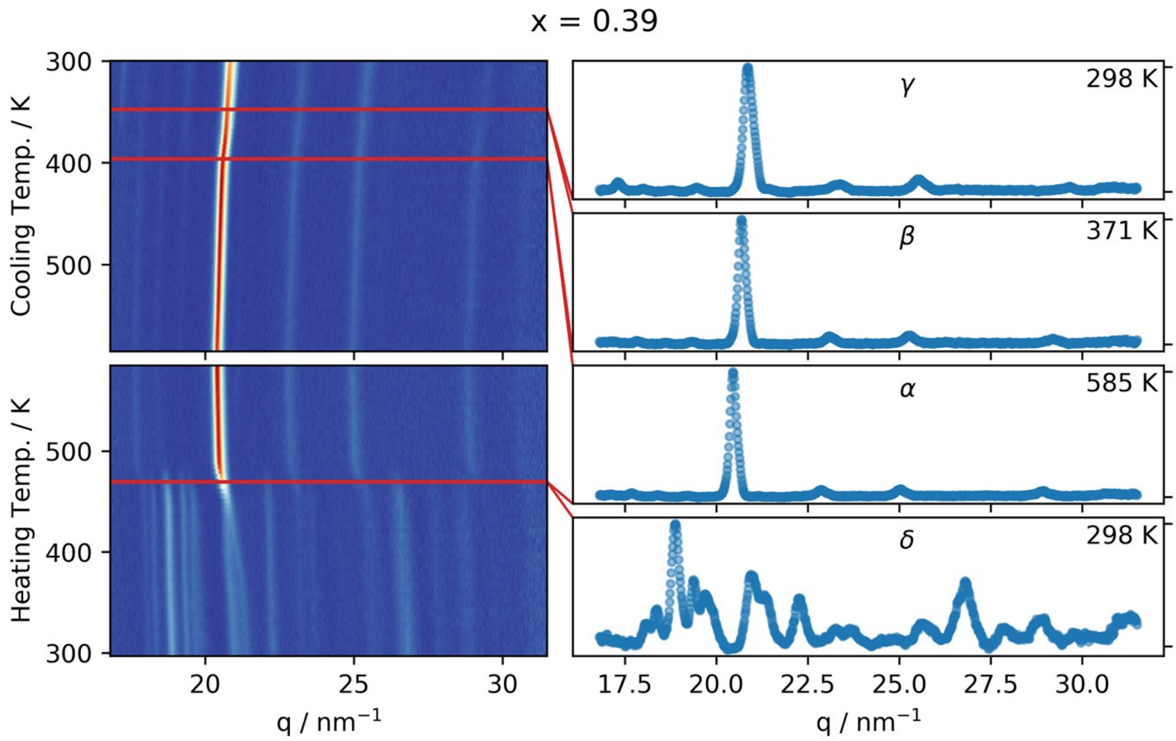

Fig. S7 Left, integrated GIWAXS pattern as a function of temperature for $CsPb(Br_xI_{1-x})_3$ with $x = 0.39$ (determined by in-situ XRF) as the sample is heated and then cooled at a rate of 2.5 K/min. Red lines indicate the phase transitions. Right, integrated GIWAXS patterns for the four different phases in the sample.

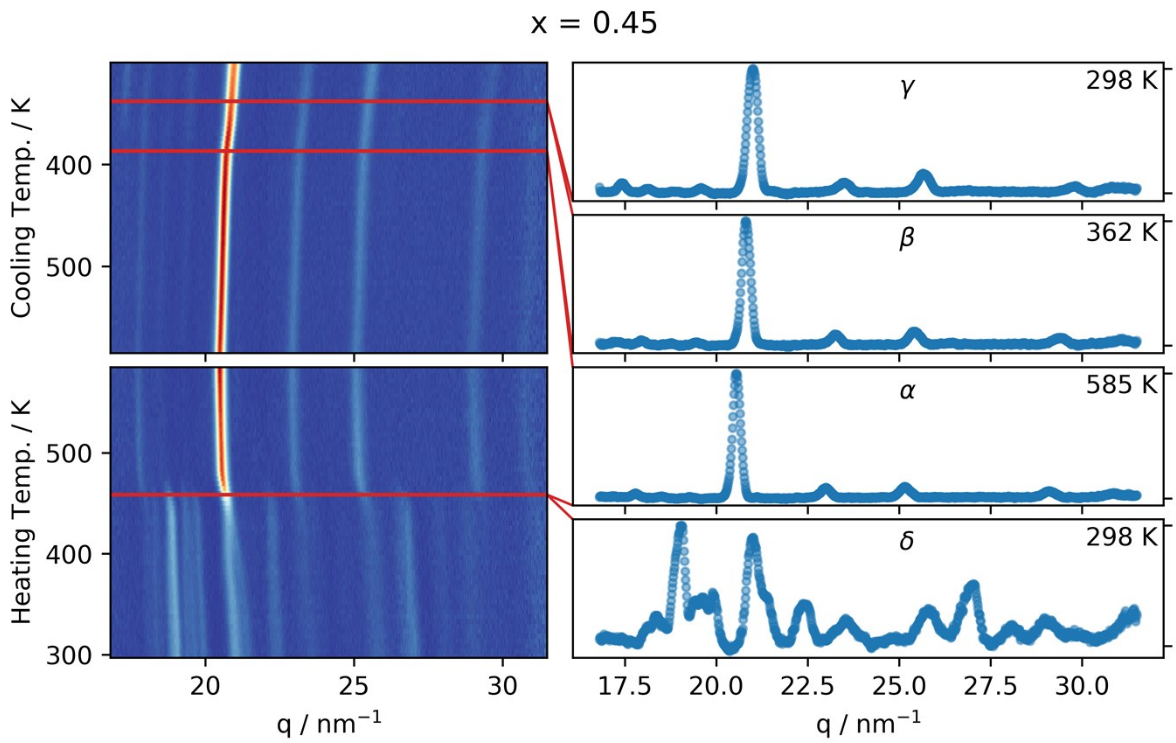

Fig. S8 Left, integrated GIWAXS pattern as a function of temperature for $CsPb(Br_xI_{1-x})_3$ with $x = 0.45$ (determined by in-situ XRF) as the sample is heated and then cooled at a rate of 2.5 K/min. Red lines indicate the phase transitions. Right, integrated GIWAXS patterns for the four different phases in the sample.

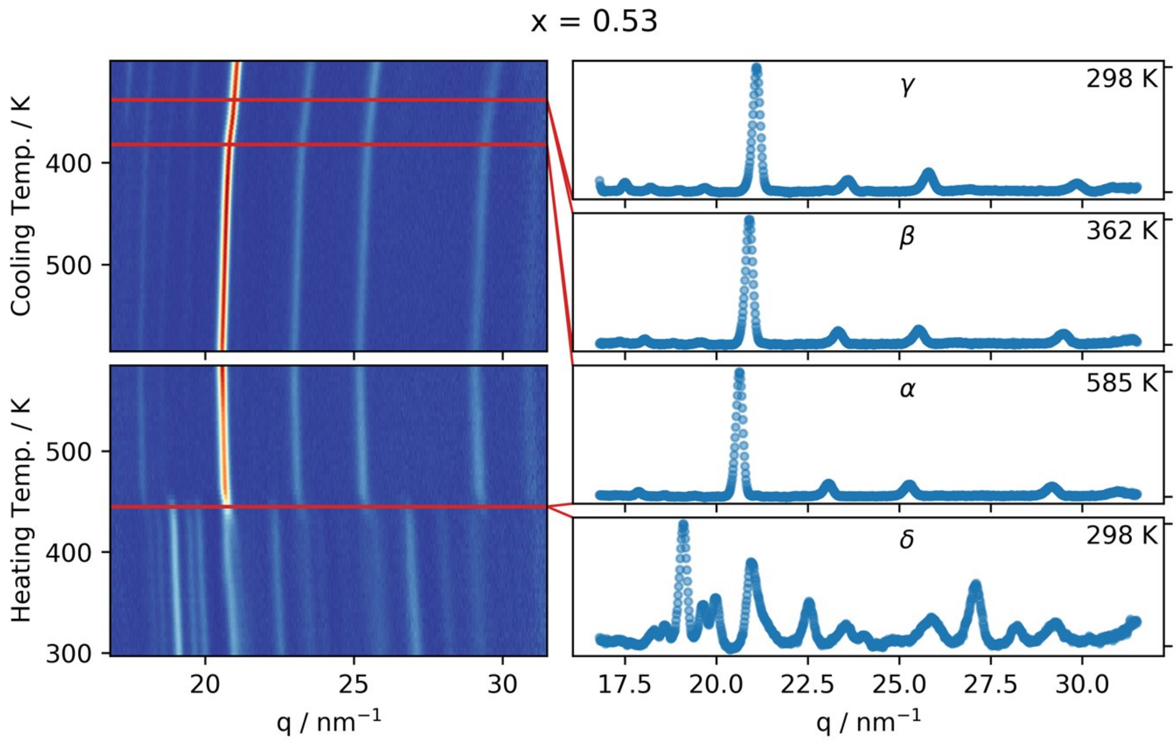

Fig. S9 Left, integrated GIWAXS pattern as a function of temperature for $CsPb(Br_xI_{1-x})_3$ with $x = 0.53$ (determined by in-situ XRF) as the sample is heated and then cooled at a rate of 2.5 K/min. Red lines indicate the phase transitions. Right, integrated GIWAXS patterns for the four different phases in the sample.

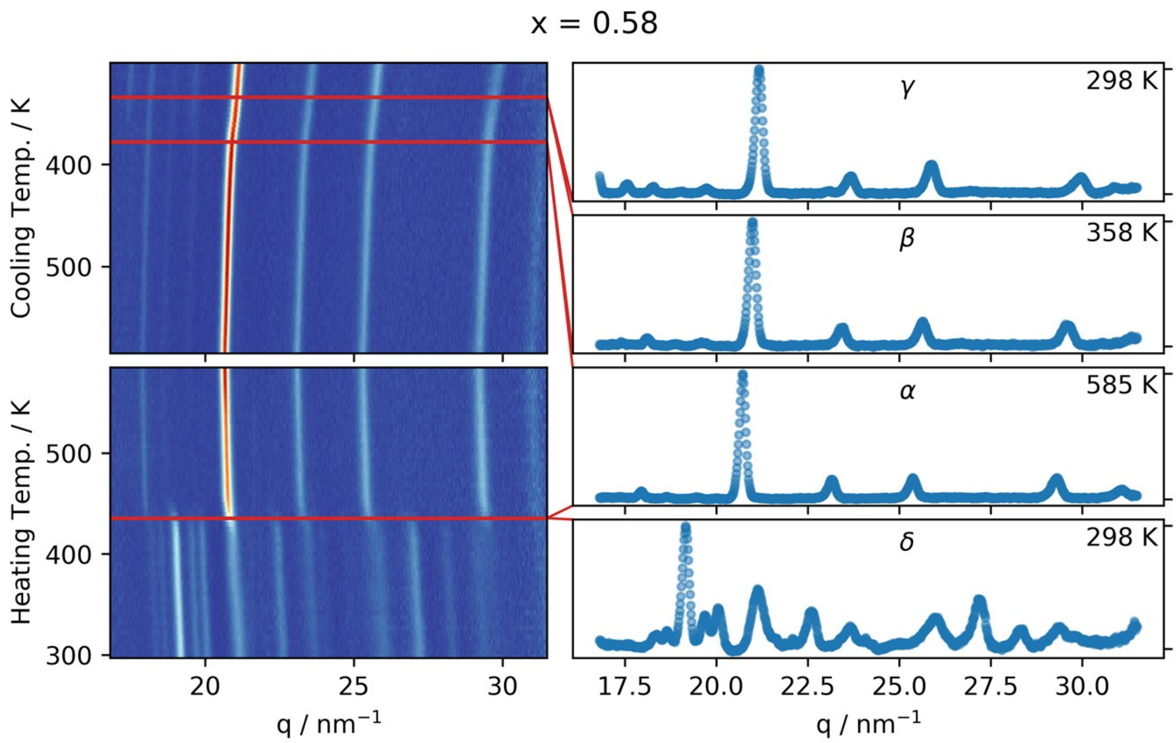

Fig. S10 Left, integrated GIWAXS pattern as a function of temperature for $CsPb(Br_xI_{1-x})_3$ with $x = 0.58$ (determined by in-situ XRF) as the sample is heated and then cooled at a rate of 2.5 K/min. Red lines indicate the phase transitions. Right, integrated GIWAXS patterns for the four different phases in the sample.

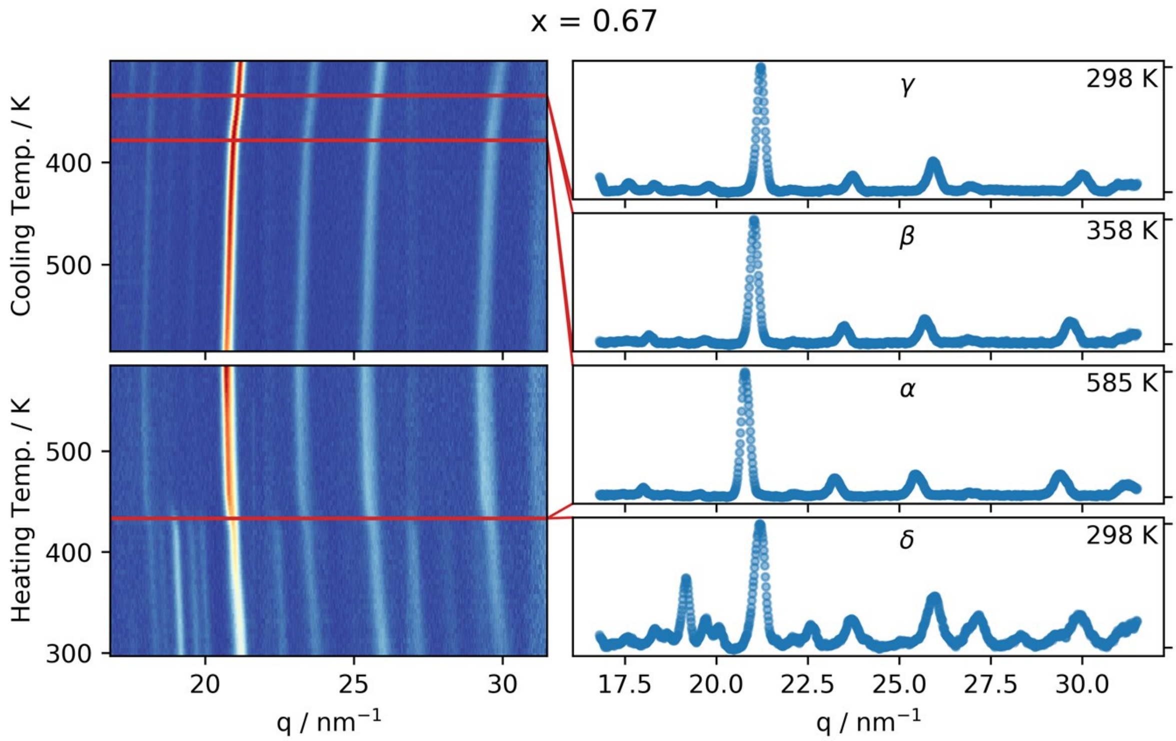

Fig. S11 Left, integrated GIWAXS pattern as a function of temperature for $CsPb(Br_xI_{1-x})_3$ with x = 0.67 (determined by in-situ XRF) as the sample is heated and then cooled at a rate of 2.5 K/min. Red lines indicate the phase transitions. Right, integrated GIWAXS patterns for the four different phases in the sample.

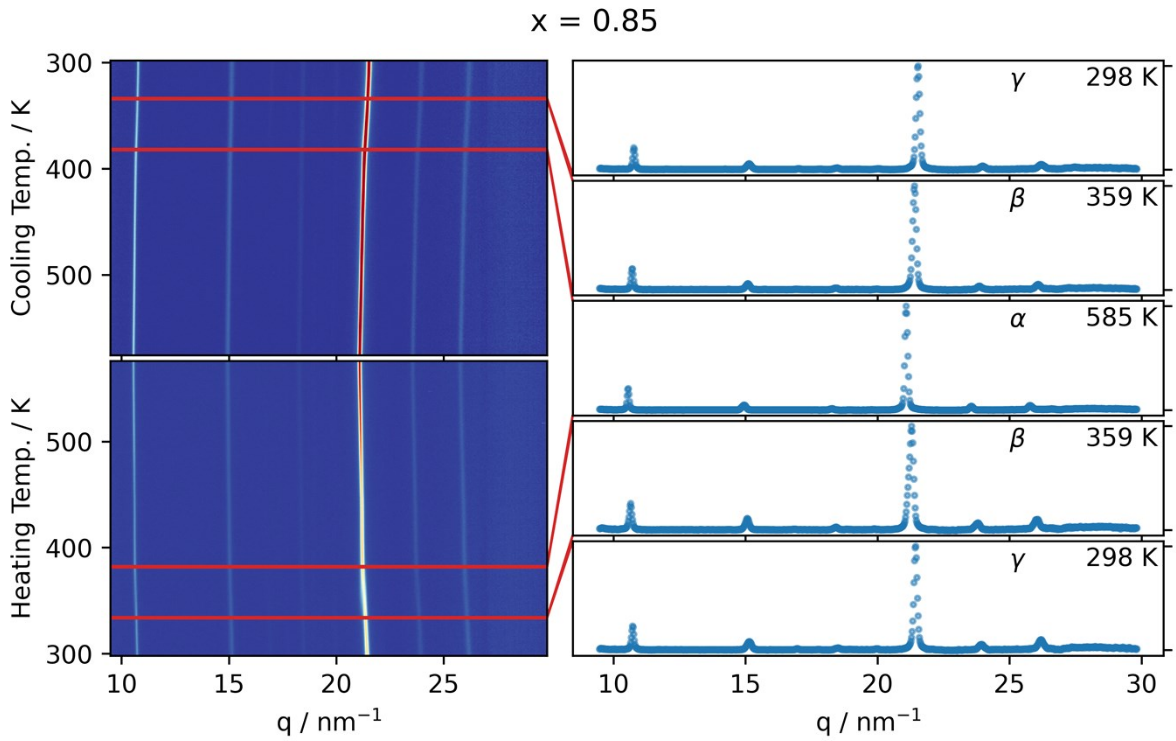

Fig. S12 Left, integrated GIWAXS pattern as a function of temperature for the spincoated $CsPb(Br_xI_{1-x})_3$ reference with x = 0.85 (determined by ex-situ XRF) as the sample is heated and then cooled at a rate of 2.5 K/min. Red lines indicate the phase transitions. Right, integrated GIWAXS patterns for the three different phases in the sample.

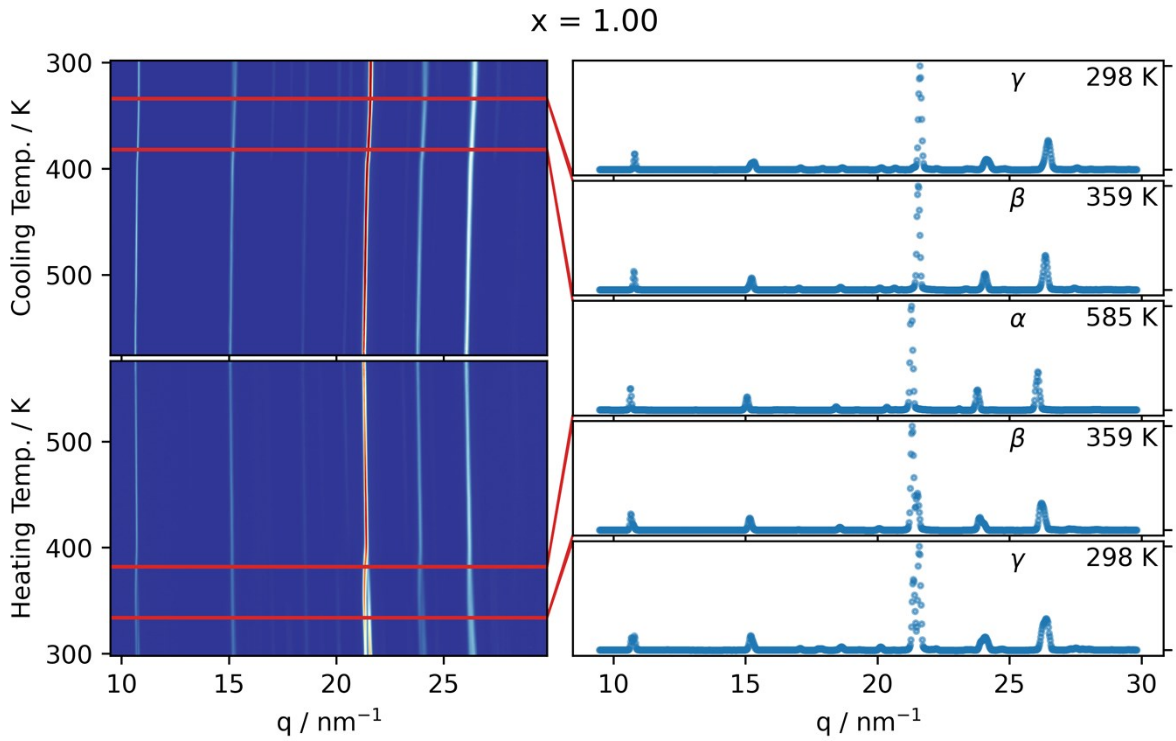

Fig. S13 Left, integrated GIWAXS pattern as a function of temperature for the blade-coated $CsPb(Br_xI_{1-x})_3$ reference with $x = 1$ as the sample is heated and then cooled at a rate of 2.5 K/min. Red lines indicate the phase transitions. Right, integrated GIWAXS patterns for the three different phases in the sample.