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## Crystallization pathway control of $\text{Bi}_2\text{Fe}_4\text{O}_9$ studied by in situ Pair Distribution Function analysis

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The targeted control of particle/crystallite size, crystallinity, and polymorphism is of crucial importance for many functional materials, in particular quantum materials<sup>1</sup>. They are frequently synthesized by the facile sol-gel method which in a broader sense can be described as the conversion of molecular precursors in solution into inorganic solids via hydrolysis, condensation, and aggregation<sup>2</sup>. The synthesis of pure nanocrystalline samples, however, can be very difficult if various stable and metastable phases exist often leading to co-crystallization. In this study, we use *in situ* total scattering and Pair Distribution Function (PDF) analysis to follow the transformation of molecular precursors into multiferroic  $\text{Bi}_2\text{Fe}_4\text{O}_9$  with a second-scale time resolution. The precursors were synthesized by the sol-gel process using the respective metal nitrates and meso-erythritol as the complexing agent. We show how the crystallization pathways and kinetics of the target compound can dramatically be changed by variation of the synthesis medium and ratio of metal nitrate to the complexing agent and relate this to the precursor structure. As an example, using small amounts of complexing agent leads to a crystalline precursor which first gets amorphous at 613 K, crystallizes into  $\text{BiFeO}_3$  at 706 K, and subsequently transforms into  $\text{Bi}_2\text{Fe}_4\text{O}_9$  at 815 K. On the other hand, bigger amounts of complexing agent produce an amorphous precursor which directly crystallizes into  $\text{Bi}_2\text{Fe}_4\text{O}_9$  at 762 K. Using PDF we reveal the importance of the initial gel structure in the overall crystallization behaviour of the system.

<sup>1</sup> Samarth, N. *Nat. Mater.*, 2017, 16, 1068.

<sup>2</sup> Niederberger, M. *Acc. Chem. Res.*, 2007, 40, 793.

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