Harmonic light scattering probes structure and defects during the fast crystallization of ZIF-8

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When it comes to its crystallization mechanism, ZIF-8 is one of the most extensively studied metal-organic frameworks (MOFs). Traditional techniques based on X-ray and light scattering have provided detailed insight into the complex pathways of nucleation and crystal growth of ZIF-8. However, especially at the initial stages, open questions remain.

Here, we probe the nucleation and crystal growth of ZIF-8 nanocrystals with in-situ nonlinear optical (NLO) light scattering techniques. NLO processes are remarkably sensitive towards symmetry and are able to characterize the structure of aggregates, clusters and crystals in-situ.1,2 Our in-situ NLO measurements provided unique insights into the crystallization mechanism of ZIF-8, which were missed by SAXS/WAXS synchrotron performed on the same samples. In the earliest stage fast nucleation and growth lead to a high concentration of defects which were probed by combining second and third harmonic light scattering. The earliest structure of the crystals is picked up by depolarised measurements and is in line with the final crystal structure of ZIF-8. In the later stages Ostwald ripening leads to defect healing increasing the overall crystallinity.

Our measurements highlight the potential of NLO techniques to probe nucleation and crystal growth in situ, providing a window into information missed by traditional X ray based techniques.

1 Van Cleuvenbergen, S. et al. Nat. Commun. 9, 3418 (2018)
2 Van Steerteghem, N. et al. Anal. Chem. 89, 2964–2971 (2017)