Structural signatures of the glass transition in emulsions

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The nature of colloidal glasses and the glass transition remains a topic of scientific interest. Scientists often focus on the study of dynamical properties since major structural changes have not been found to date in the vicinity of the glass transition. In this work we investigate structural and dynamical properties of moderately polydisperse emulsions across an extended range of droplet volume fractions ϕ , encompassing fluid and glassy states up to jamming. Combining experiments and simulations, we show that when ϕ approaches the glass transition volume fraction ϕ_g , dynamical heterogeneities and amorphous order arise within the emulsion. In particular, we find an increasing number of clusters of particles having five-fold symmetry (i.e. the so-called locally favoured structures, LFS) as ϕ approaches ϕ_g , saturating to a roughly constant value in the glassy regime. However, contrary to previous studies, we do not observe a corresponding growth of medium-range crystalline order; instead, the emergence of LFS is decoupled from the appearance of more ordered regions in our system. We also find that the static correlation lengths associated with the LFS and with the fastest particles can be successfully related to the relaxation time of the system. By contrast, this does not hold for the length associated with the orientational order. Our study reveals the existence of a link between dynamics and structure close to the glass transition even in the absence of crystalline precursors or crystallization.