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Quenching by heating: colloidal liquid-gas critical phenomena at the single particle level.

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⁰*Motivation for Oral: We have resolved a fluid-fluid interface at the single particle level in 3D, investigated critical fluctuations and the correlation length in a polymer-colloid mixture. Furthermore we exploit the temperature dependence of the size of the polymer depletion agent to induce phase separation in-situ. To our knowledge this is the first time either has been demonstrated. We believe that temperature-induced demixing can further the investigation of phase separation in its earliest stages, as intrinsically unstable samples usually begin phase separating before observation. Our single particle analysis is an ideal tool for studying the earliest stages of phase separation. Finally we provide an intriguing demonstration of a phase separated system which we can redisperse, or 'boil', by cooling.*

We present two significant advances in the familiar depletion mechanism of 'liquid-gas' phase separation in colloid-polymer mixtures: single-particle coordinate tracking and in-situ control of the effective temperature. We use the colloidal model system of fluorescently labelled poly(methyl methacrylate), in a closely density-matched and refractive index-matched solvent, with polystyrene (PS) whose osmotic pressure induces depletion attractions between the colloids. We use colloids of the micron length-scale to facilitate single-particle tracking with confocal microscopy, and back up this technique with dynamic light scattering, which accesses a macroscopic sample volume.

Since PS is just above its theta temperature, we find relatively small changes in temperature, around 10°C, enhance the excluded volume effect sufficiently to change the polymer radius of gyration by up to 20%. This translates into a very considerable change in polymer 'volume fraction', and consequently osmotic pressure, increasing the depletion interaction between the colloids. This means that by gentle heating, we can induce phase separation in-situ, and so observe the very early linear Cahn-Hilliard regime at the single particle level. Usually this

is hard to access, since intrinsically unstable samples begin phase separating before observation.

By cooling, we find that phase separation is even reversible, which may have practical consequences, as colloid-polymer mixtures are found frequently in industrial and consumer products, and phase separation has implications for shelf-life. Thus we demonstrate the paradoxical behaviour that heating induces quenching, while cooling causes 'boiling'.

Our first study with this system explores critical phenomena on either side of the critical point, in particular the correlation length. We compare critical and off-critical fluctuations. The single particle resolution enables direct quantitative determination of the bulk correlation length in the single phase region, and in both the dense and dilute phases in the phase separated regime. We obtain a further measure of the correlation length by analyzing the *intrinsic* liquid-gas interfacial width at the single particle level. We find good agreement between the various methods.