ID de Contribution: **213** Type: **Contribution orale**

A double rigidity transition rules the fate of drying colloidal drops

lundi 3 juillet 2023 16:45 (27 minutes)

The evaporation of drops of colloidal suspensions plays an important role in numerous contexts, such as the production of powdered dairies, the synthesis of functional supraparticles, and virus and bacteria survival in aerosols or drops on surfaces. The presence of colloidal particles in the evaporating drop eventually leads to the formation of a dense shell that may undergo a shape instability. Previous works propose that, for drops evaporating very fast, the instability occurs when the particles form a rigid porous solid, constituted of permanently aggregated particles at random close packing. To date, however, no measurements could directly test this scenario and assess whether it also applies to drops drying at lower evaporation rates, severely limiting our understanding of this phenomenon and the possibility of harnessing it in applications. Here, we combine macroscopic imaging and space- and time-resolved measurements of the microscopic dynamics of colloidal nanoparticles in drying drops, measuring the evolution of the thickness of the shell and the spatial distribution and mobility of the nanoparticles. We find that, above a threshold evaporation rate, the drop undergoes successively two distinct shape instabilities. While the second instability is due to the permanent aggregation of nanoparticles, as hypothesized in previous works on fast-evaporating drops, we show that the first one results from a reversible glass transition of the shell, unreported so far. We rationalize our findings and discuss their implications in the framework of a unified state diagram for the drying of colloidal drops.

Affiliation de l'auteur principal

Laboratoire Charles Coulomb, CNRS et Université de Montpellier

Auteurs principaux: MILANI, Matteo (Laboratoire Charles Coulomb, CNRS et Université de Montpellier); Dr PHOU, Ty (Laboratoire Charles Coulomb, CNRS et Université de Montpellier); Prof. LIGOURE, Christian ((Laboratoire Charles Coulomb, CNRS et Université de Montpellier); Prof. CIPELLETTI, Luca (Laboratoire Charles Coulomb, CNRS et Université de Montpellier); RAMOS, laurence (Laboratoire Charles Coulomb, CNRS et Université de Montpellier)

Orateur: RAMOS, laurence (Laboratoire Charles Coulomb, CNRS et Université de Montpellier)

Classification de Session: Mini-colloques: MC15 Matière molle : des concepts fondamentaux à la fabrication de systèmes originaux

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