Highlights

Physics of Materials —2017

Cage Size and Jump Precursors in Glass-Forming Liquids: Experiment and Simulations

R. Pastore[1,2], G. Pesce[3], A. Sasso[3], M. Pica Ciamarra[4,2]

[1] Dipartimento di Ingegneria Chimica, dei Materiali e della Produzione Industriale, Universitá di Napoli Federico II, Napoli, Italia
[2]CNR-SPIN, Napoli, Italia
[3]Dipartimento di Fisica, Università di Napoli Federico II, Napoli, Italia
[4]Division of Physics and Applied Physics, Nanyang Technological University, Singapore

The Journal of Physical Chemistry Letters 8, 1562 (2017)

The relaxation dynamics of glassy systems proceeds via a sequence of sudden jumps, through which a particle changes its neighbors (Fig.1). These jumps are not homogeneously distributed neither in time nor in space. Relating the resulting heterogeneous particle dynamics to the diverse structural environments explored by the particles is a notorious difficult task, but crucial to rationalize the relaxation process. In this paper, we have introduced a novel approach that takes advantage of the cage-jump events in the single-particle trajectories. We use the cage size as a proxy of the local structure and the jump as a proxy of the local relaxation. Both simulations of supercooled liquids and experiments on hard-sphere colloidal suspensions show that a cage opening process takes place shortly before the jump, whereas the cage size attains a plateau at larger time. The plateau value is smaller for longer-lasting cages, revealing a clear coupling between local structure and dynamics (Fig.2). We clarify how this coupling control the macroscopic behaviour of soft glassy materials and opens the way to a better understanding of their relaxation process.

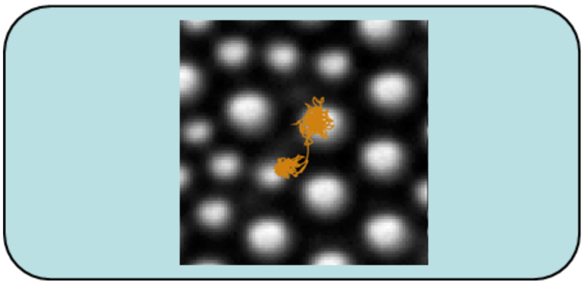

Fig.1: The trajectory of a particle of a dense colloidal assembly reveal the presence of a cage-jump .

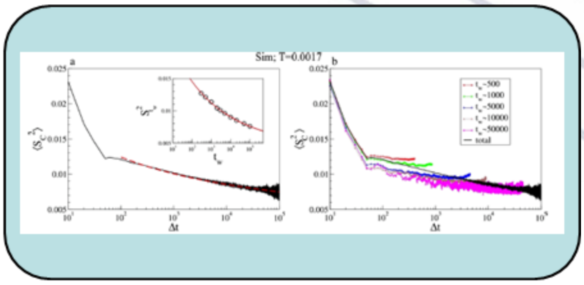

Fig.2: From simulations: (a) cage size averaged over the total ensemble of cages (solid line). At long time, data are correctly approximated by our model (dashed line) (b) Cage size averaged over the total ensemble (line) and over subensembles of cages of fixed waiting time (line-points). (Inset) Long time plateau of the cage size.