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### Direct Observation of Percolation in the Yielding Transition of Colloidal Glasses

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# Supplemental Material for article "Direct observation of percolation in the yielding transition of colloidal glasses"

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## I. COMPARISON WITH OSCILLATORY "YIELDING"

Besides the linear strain ramp (constant strain rate) applied in the main manuscript, recent studies, in particular of soft glasses, include oscillatory probing, in which the sample is subjected to oscillatory strain, typically of increasing amplitude. The elastic-to-plastic transition then occurs from the small-amplitude linear to the large-amplitude nonlinear regime. While this transition has been referred to as "yielding" as well, we stress that this protocol is distinct and probes a different state of the material: the linear strain ramp approaches steady state flow at constant strain rate, while the latter approaches a steady oscillatory state. In material science, the term "yielding" has been traditionally associated with the material response to a linear strain ramp in the sense we use it, while recent literature on soft glasses has extended this term to the oscillatory probing [2], where it is loosely associated with either the strain at which the storage modulus starts to deviate from its small-strain amplitude value or the point at which the storage and loss moduli cross over, or any point in between. We believe the two shear protocols (linear strain ramp and oscillatory shear) are fundamentally different and thus not straight forward to compare. Most notably, the oscillatory protocol subjects the material to sinusoidal strain of not only increasing amplitude, but also increasing strain-rate amplitude [34]. This different shear protocol may lead to a different yielding scenario, as suggested by recent work, both experiments and simulations: a first-order like scenario under oscillatory shear [7,35-38] and a critical or second-order like scenario at constant strain rate as in our case. Interestingly, strain-ramp experiments conducted at increasing strain rate exhibited a first-order like transition

as well [39], suggesting that it is the varying *strain rate* that leads to a discontinuous transition. While this needs further investigation, one can still try to compare yield strains under both protocols. A possible way to map both protocols has been suggested by Rogers *et al.* [40], and applied to colloidal glasses by van der Vaart *et al.* [41]: by regarding each oscillation cycle as consisting of straining and yielding, one can extract yield stresses as a function of strain amplitude in a way similar to continuous straining, see Fig. S1. The hereby extracted yield strain of around 0.08 for a volume fraction of  $\phi = 0.61$  close to our value of 0.6 (see Fig. S1b) is close to the crossing point of  $G'$  and  $G''$ , which is between 0.07 and 0.08 for the same volume fraction (see Fig. S1a). These values are indeed consistent with the yield strain of  $\sim 0.09$  observed in the present manuscript, showing that the yield strain may correspond to the crossing of  $G'$  and  $G''$ . This crossing is also the point where a sharp first-order-like transition is observed in oscillatory measurements [35]. But we do think that the protocols are fundamentally different, and therefore, this comparison should merely serve as a guideline.

## II. INFLUENCE OF STRAIN RATE IN ATOMISTIC SIMULATIONS

In addition to the simulations at strain rate  $2 \times 10^8 \text{ s}^{-1}$  reported in the manuscript, we also performed atomistic simulations at strain rates two and five times higher. The resultant cluster sizes and correlation length for the different strain rates are quantitatively similar as shown in Fig. S2. Obviously, the strain rate does not change the qualitative behavior, and has only minor quantitative effects on the results. Consequently, we observe a very similar percolation transition in all cases, irrespective of the strain rate.

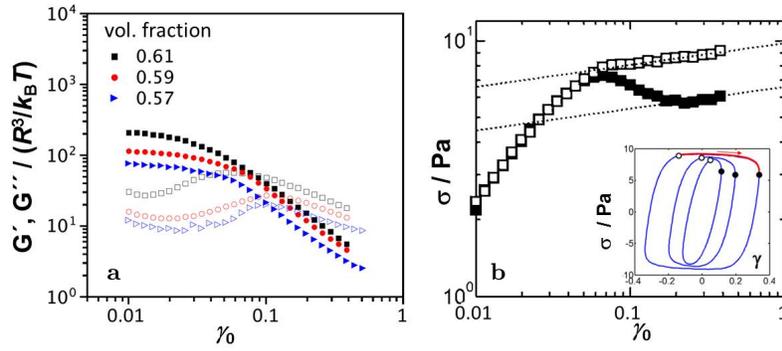


FIG. S1. **Oscillatory rheology of hard-sphere colloidal suspensions.** (a) Storage and loss moduli for different volume fractions at frequency  $1 \text{ s}^{-1}$ . (b) Stress as a function of strain amplitude at the upper and lower yield points (see Lissajous curves in the inset, open and closed symbols, respectively) within each oscillation cycle for volume fraction  $\phi = 0.61$  corresponding to the black data in panel (a). In both cases, yielding occurs at strains of around 7-8 %, which is also very similar to the microscopic yield strain of 9% reported in the main manuscript.

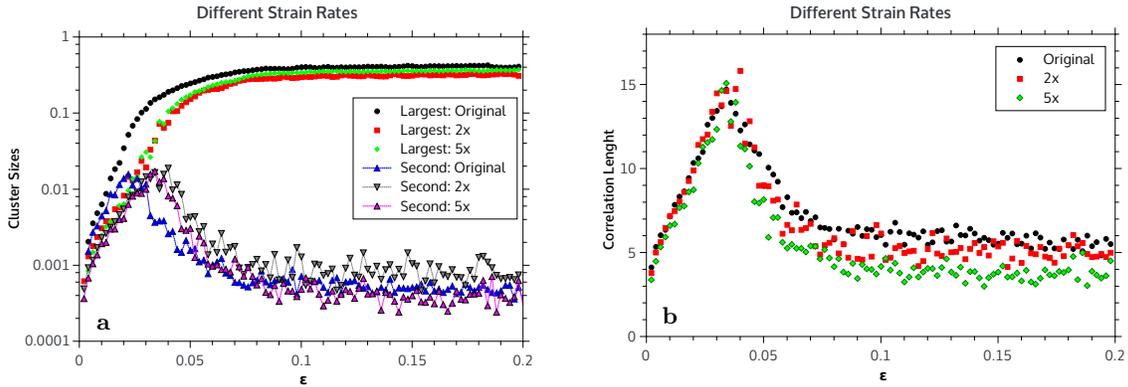


FIG. S2. **Influence of strain rate in atomistic simulations.** (a) Largest and second largest cluster as a function of strain for different strain rates: original strain rate ( $\dot{\gamma} = 2 \times 10^8 \text{ s}^{-1}$ ), twice ( $4 \times 10^8 \text{ s}^{-1}$ ) and five times higher ( $10 \times 10^8 \text{ s}^{-1}$ ). (b) Correlation length as a function of strain for the same strain rates. The qualitative behavior is robust with strain rate changes; only minor quantitative differences are observed.