Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information: Homogeneous Nucleation of Sheared Liquids: Advances and Insights from Simulations and Theory

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1 Nucleation Parameters

1.1 Computational Approaches

Table S1 summarizes the range of shear rates considered and calculated nucleation rates, J, for water, using brute-force approaches and the shear-CNT formalism. We note that the CNT-based formalisms of Mura et al.[S3] and Goswami et al.[S2,S5] can, in principle, accept any positive value of $\dot{\gamma}$. However, in practice, the shear rate range to be analysed using these CNT-based approaches can be chosen to pinpoint the non-monotonic regime of J, such that $\dot{\gamma} < \dot{\gamma}_{\text{max}}$.[S5] Table S2 lists the values of supersaturation and shear rate ranges considered in selected recent studies of hard-sphere (HS) colloids.

Table S1: Orders of magnitudes of the calculated nucleation rates, J ($\mathrm{m}^{-3}\mathrm{s}^{-1}$), and orders of magnitudes of the approximate range of shear rates, $\dot{\gamma}$ (s^{-1}), for which non-monotonicity in J is observed, for different water models at various supercoolings, ΔT (K). Here, MFPT and BF refer to the mean first-passage time method (Section 2.1.2 in the main text) and brute-force approaches in which J is calculated according to Eq. 7 in the manuscript, respectively.

Model	**Method**	**ΔT**	**Order of $\dot{\gamma}$**	**Order of J**	**References**
mW	BF	67.6	$8-9$	$31-32$	Luo et al.[S1]
	MFPT			$36-37$	
	shear-CNT	14.6	8	-185	Goswami et al.[S2]
		19.6	$8-9$	-82	
		34.6	9	-3	
		39.6	9	6	
TIP4P/2005	shear-CNT	14.5	$6-7$	-186	Goswami et al.[S2]
		19.5	7	-82	
		29.5	7	4	
TIP4P/Ice	shear-CNT	14.5	7	-174	Goswami et al.[S2]
		19.5	7	-75	
		34.5	7	0	

Table S2: Values of the packing fraction, ϕ and ranges of shear rates, $\dot{\gamma}$, considered in the application of different methods for the analysis of hard sphere (HS) colloids. CNT refers to the CNT-based approach of Mura et al.[S3] (described in Section of 2.3.1 of the main text) and τ_B is the Brownian time (which should not be confused with the characteristic time τ defined in Section 2.3.1). Non-monotonicity in the sheared nucleation rates was reported by Mura et al.[S3] for the approximate shear rate ranges provided here (in powers of 10).

Method	**ϕ**	**$\dot{\gamma}$**	**References**
BF	$0.539-0.587$	$\leq 1.414\tau_B^{-1}$	Richard et al.[S4]
CNT	0.06	$(10^{-3}-10^{-2})\ \mathrm{s}^{-1}$	Mura et al.[S3]
	0.15	$10^{-3}\ \mathrm{s}^{-1}$	
	0.503	$(10^{-4}-10^{-3})\ \mathrm{s}^{-1}$	

1.2 Experiments

Table S3: Orders of magnitude of the shear rates, $\dot{\gamma}$ (s^{-1}), generated in protein systems, via various capillary and rotational devices.[S6]

Device type	**Order of $\dot{\gamma}$**	**References**
Couette cell	3	Singh et al.[S7]
Taylor-Couette flow system	1	Ashton et al.[S8]
Capillary setup	$4-5$	Thomas et al.,[S9] Jaspe et al.[S10]
Microfluidic cell	3	Schneider et al.[S11]

Table S3 lists the orders of magnitude of shear rates imposed in protein systems, using different experimental techniques. For comparison with theoretical and computational approaches, generating a uniform flow field (or in other words, non turbulent flow) is desirable. As noted in Section 4 in the main text, imposing a uniform flow field in liquids such as supercooled water is challenging. It is also evident that the shear rates of interest for water, as indicated by computational techniques (Table S1), tend to be orders of magnitudes greater than the shear rates that can be typically generated in experiments.

On the other hand, shear rate ranges for which non-monotonicity in J is theoretically observed in colloidal suspensions (Table S2) are accessible by experimental techniques (Table S3).

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