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# A universal scaling framework for controlling phase behavior in thickening and jamming suspensions

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Recently, we proposed a universal scaling framework that shows shear thickening in dense suspensions is governed by the crossover between two critical points: one associated with frictionless isotropic jamming and a second corresponding to frictional shear jamming. Here, we show that orthogonal perturbations to the flows, an effective method for tuning shear thickening, can also be folded into this universal scaling framework. Specifically we show that the effect of adding orthogonal shear perturbations (OSP) can be incorporated into the scaling variable via a multiplicative function, determined through our measurements, to achieve collapse of the entire thickening and dethickening data set onto a single universal curve. We then show that this universal scaling framework can be used to control the phase behavior in thickening and jamming suspensions.

Shear thickening suspensions demonstrate an increase in the viscosity upon increasing the suspension stress [1, 2]. This increase in the viscosity has been attributed to the change in the nature of contacts from lubrication to frictional [3–7] and can be very large and even lead to jamming at sufficiently high packing fractions. We recently proposed a scaling formulation that describes the shear thickening suspension viscosity as crossover between behaviours governing two different critical points. At small applied stresses, the suspension viscosity is determined by the distance to the frictionless isotropic jamming critical point. As the shear stress is increased and frictional contacts form between the particles, the flow properties transition to a new universal behavior, governed by the distance to the frictional shear jamming critical point, which occurs at lower volume fractions [8]. Interestingly, a growing body of work is showing that incorporating additional multi-directional flows can be used to modify the contact network and tune the suspension viscosity [9–16]. Whether these more complex flows can be incorporated into a universal scaling framework is yet to be explored.

Here, we investigate how tunability via orthogonal superimposed perturbations (OSP) modifies the scaling relations. Orthogonal superimposed perturbations have been used in a range of colloidal systems from gels to glasses and dispersions to both probe and tune the system rheology [9, 11, 17–22]. In colloidal gels and glasses, it has been shown that by applying small amplitude sinusoidal flows orthogonal to the primary shear flow, one can measure the time dependent viscoelastic response of structures formed under shear [17–20]. In shear thickening suspensions OSP flows can be used as tuning knob – these orthogonal flows can break up the fragile force chains that lead to thickening and alter the suspension viscosity [9, 11]. Importantly, applying the scaling analysis to OSP flows, and establishing universal principles requires extensive data collected over the broadest range of parameters possible. While previous OSP experiments

clearly demonstrated the ability to tune the suspension viscosity, they were only conducted at one volume fraction and one shear rate [9]. In this work, we perform experiments on a cornstarch in glycerol suspension for seven volume fractions ranging from  $\phi = 0.37$  to  $\phi = 0.52$  and for shear rates corresponding to the entire thickening region.

The average viscosity in the primary flow direction is measured with an ARES G2 rheometer, using a double wall Couette geometry. The primary flow is generated by the continuous rotation of the outer cup about the vertical  $z$  axis and the orthogonal flow is generated by the vertical oscillation of the inner bob. A steady primary flow rate ( $\dot{\gamma}_P$ ) at sufficiently large imposed strain rates results in formation of force chains and thickening of the suspension. The resulting viscosity is tuned by superimposing a small amplitude,  $\gamma_{OSP} \leq 0.05$ , sinusoidal orthogonal flow.

The viscosity measurements are shown in Fig. 1(a) and (b). In the absence of any orthogonal flows we observe typical thickening behavior (Fig. 1(a)). Here, each volume fraction is indicated with a different symbol, and each primary strain rate is indicated with a different color (Fig. 1 side bar). The effect of OSP flow on the suspension viscosity at one volume fraction,  $\phi = 0.44$ , is shown in Fig. 1(b). At each primary shear rate, OSP flow is applied with a fixed amplitude  $\gamma_{OSP} = 0.05$ , and angular frequencies,  $\omega_{OSP}$ , ranging from  $0.05 s^{-1}$  to  $100 s^{-1}$ . The suspension response is dependent on both the primary and the orthogonal flows (Fig. 1(b), inset). At large primary shear rates, we observe no changes in the viscosity for all applied OSP flow rates. At intermediate primary shear rates, we observe dethickening at sufficiently high OSP flows. At small primary shear rates, we observe minimal decrease in viscosity. To untangle the complex behaviour observed upon the application of OSP flows, we note that the viscosity in suspensions with OSP flows is expected to be governed by the ratio of the force chain disruption to force chain formation, which in

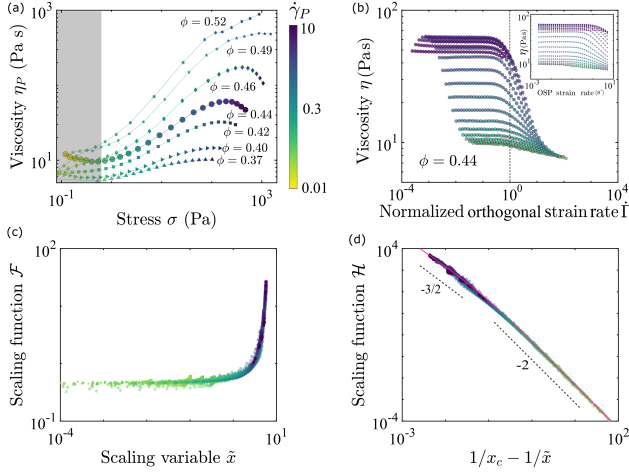


FIG. 1. **Viscosity upon application of primary and orthogonal superposition shear and scaling collapse**

(a) The viscosity versus primary stress for suspensions with volume fractions from  $\phi = 0.37$  to  $\phi = 0.52$  with no OSP flow. The primary stress is calculated as the product of the applied primary strain rate and measured primary viscosity,  $\sigma = \dot{\gamma}_P \eta_P$ . Colors correspond to different primary strain rates, ranging from  $0.01 s^{-1}$  (yellow) to  $10 s^{-1}$  (dark blue). The shaded grey region indicates the shear thinning regime and is not included in the scaling collapse. (b) The viscosity versus normalized orthogonal strain rate  $\dot{\Gamma} = \gamma_{OSP} \omega_{OSP} / \dot{\gamma}_P$  for  $\phi = 0.44$ . Primary strain rates range from  $0.01 s^{-1}$  (yellow) to  $10 s^{-1}$  (dark blue). The OSP strain amplitude of  $\gamma_{OSP} = 0.05$ . The OSP frequency,  $\omega_{OSP}$ , is varied from  $0.05 s^{-1}$  to  $100 s^{-1}$ . The decrease in viscosity occurs at  $\dot{\Gamma} \approx 1$  for all primary strain rates (dashed grey line). Inset shows the same data versus the OSP strain rate. (c) The scaling function  $\mathcal{F} = \eta(\phi_0 - \phi)^2$  versus the scaling variable,  $\tilde{x}(\sigma, \dot{\Gamma}, \phi) = f(\sigma)g(\dot{\Gamma})C(\phi)/(\phi_0 - \phi)$  where  $f(\sigma) = e^{-(\sigma^*/\sigma)^{0.75}}$  and  $g(\dot{\Gamma}) = e^{-\dot{\Gamma}/\dot{\Gamma}^*}$  showing excellent collapse onto a universal curve that diverges at  $\tilde{x} = x_c$ . (d) The scaling function  $\mathcal{H} = \eta(\zeta(\sigma, \dot{\Gamma}, \phi))^2$  versus  $|1/x_c - 1/\tilde{x}|$  where  $\zeta(\sigma, \dot{\Gamma}, \phi) = f(\sigma)g(\dot{\Gamma})C(\phi)$  showing 2 distinct power law regimes - frictionless jamming  $\tilde{x}^{-2}$  and frictional jamming  $\sim \tilde{x}^{-1.5}$  (dashed grey lines). Pink line is the scaling function from [8].

its simplest form is captured by the dimensionless strain rate  $\dot{\Gamma} = \gamma_{OSP} \omega_{OSP} / \dot{\gamma}_P$  [30] (Fig 1b). Consistent with this hypothesis, we find that the viscosity starts decreasing at  $\dot{\Gamma} \approx 1$  for all primary shear rates. The wide range of viscosities observed makes this OSP protocol an ideal first step to investigate the use of the scaling formulation in multi-directional flows.

The scaling theory [8] in the absence of multi-directional flows uses the collapse of experimental data to write the viscosity as a function of a single scaling variable,  $x_1 = \frac{f(\sigma)C(\phi)}{(\phi_0 - \phi)}$ , that includes a stress dependent fraction of frictional contacts,  $f(\sigma)$ , a multiplicative factor  $C(\phi)$  that depends on the volume fraction,  $\phi$ , and

the distance to the frictionless isotropic jamming point  $\phi_0 - \phi$ .

$$\eta(\phi_0 - \phi)^2 = \mathcal{F}(x_1) \quad (1)$$

At small  $x_1$ , the scaling function,  $\mathcal{F}$ , is constant and the viscosity diverges as  $(\phi_0 - \phi)^{-2}$ , associated with isotropic frictionless jamming. At larger values of  $x_1$  this universal function  $\mathcal{F}$  was found to have a divergence associated with frictional shear jamming. Here, the viscosity increased as  $(x_c - x_1)^{-\delta}$ , with  $\delta \sim 3/2$  [31]. The different exponents indicate that frictional shear jamming and frictionless isotropic jamming are associated with different universality classes.

Inspired by our previous work, we extend the scaling framework by introducing a new scaling variable that contains a product of independent functions that depend on the stress  $\sigma$ , dimensionless strain rate,  $\dot{\Gamma}$ , and volume fraction,  $\phi$ . We find that this approach works extremely well, collapsing our entire data set (Fig. 1(c)). Similar to the previous analysis, we express the viscosity as

$$\eta(\phi_0 - \phi)^2 = \mathcal{F}(\tilde{x}) \quad (2)$$

but this time with

$$\tilde{x}(\sigma, \dot{\Gamma}, \phi) = \frac{\zeta(\sigma, \dot{\Gamma}, \phi)}{(\phi_0 - \phi)} = \frac{f(\sigma)g(\dot{\Gamma})C(\phi)}{(\phi_0 - \phi)} \quad (3)$$

where the second equality captures a conjecture that  $\zeta$  can be expressed as a product of three independent functions. The inclusion of the multiplicative term,  $g(\dot{\Gamma})$  is similar to those used for introducing additional constraints [25] or a minimum strain [13] required for thickening. Impressively, this approach enables scaling collapse of the data across all volume fractions, stresses and OSP flow rates. In addition to demonstrating the incredible versatility our scaling framework, this scaling collapse of the data illustrates that orthogonal flows do not alter the nature of the critical points associated with the thickening transition.

Key to the scaling collapse of the viscosity is determining the scaling variable  $\tilde{x}$ . To determine the functions  $C(\phi)$ ,  $g(\dot{\Gamma})$  and  $f(\sigma)$ , we use an iterative process. We start with our previous form for  $f(\sigma) = e^{-\sigma^*/\sigma}$ , where the stress used to calculate  $f(\sigma)$  is always the primary stress,  $\sigma = \eta_P \times \dot{\gamma}_P$ . Using the scaling collapse of  $\dot{\Gamma} = 0$  data across different volume fractions, we estimate  $C(\phi)$ . With this function  $C(\phi)$  and the initial  $f(\sigma)$ , we determine the values of  $g(\dot{\Gamma})$  that best collapses the entire data set. Next, holding  $g(\dot{\Gamma})$  and  $C(\phi)$  fixed, we find the new values of  $f(\sigma)$  that optimizes scaling collapse. This process of holding two of the functions  $f(\sigma)$ ,  $g(\dot{\Gamma})$ , and  $C(\phi)$ , fixed to determine the third is repeated cyclically until a good scaling collapse of the data is achieved (see SI for intermediate scaling collapses and more details). The final functions are shown in Fig. 2. We find that the best collapse is obtained when the stress dependence is approximated by a stretched exponential  $f(\sigma) = e^{-(\sigma^*/\sigma)^{0.75}}$ ,

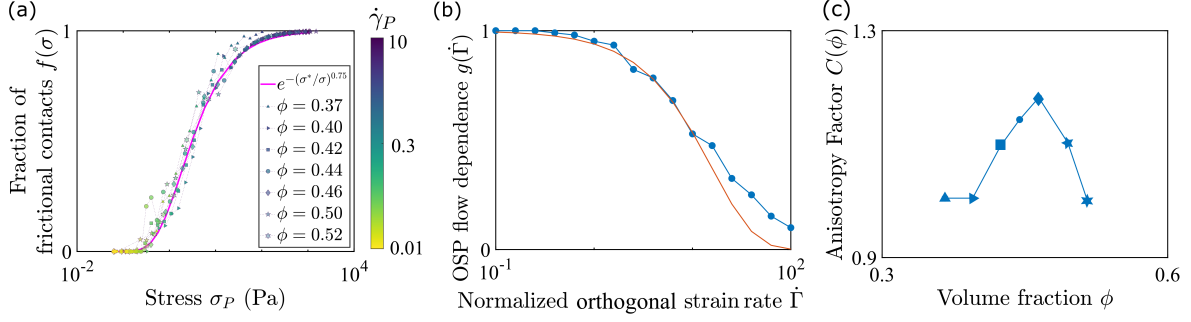


FIG. 2. **Final functions in the scaling variable** (a)  $f(\sigma)$  as a function of the applied stress. The symbols are the point by point values of  $f(\sigma)$  for each volume fraction that best collapses the data. The solid pink line is the stretched exponential form  $f(\sigma) = e^{-(\sigma^*/\sigma)^{0.75}}$  that captures the point by point data well, and is used for the best scaling collapse of the data. (b) Multiplicative function of  $\dot{\Gamma}$ ,  $g(\dot{\Gamma})$ , used to collapse the data. The blue data is the point by point value for each  $\dot{\Gamma}$  that best collapses the data. The red line is an exponential function of the normalized orthogonal strain rate  $e^{-\dot{\Gamma}/\dot{\Gamma}^*}$  that captures the transition in  $g(\dot{\Gamma})$  with increasing  $\dot{\Gamma}$ . (c) The anisotropy factor  $C(\phi)$  used for the scaling collapse of the data.

the dependence on orthogonal shear is approximated by an exponential  $g(\dot{\Gamma}) = e^{-\dot{\Gamma}/\dot{\Gamma}^*}$  and the dependence on volume fraction is non-monotonic (Fig. 2). This non-monotonic function,  $C(\phi)$  modulates the effect of  $f(\sigma)$  and  $g(\dot{\Gamma})$  across different volume fractions. While the exact origin of  $C(\phi)$  is unclear, we hypothesize that  $C(\phi)$  is related to the anisotropy or the connectivity of the force network. Such a hypothesis could indicate that the peak in  $C(\phi)$  is related to the emergence of the first rigid clusters, which is shown to occur before discontinuous shear thickening. The three functions  $f(\sigma)$ ,  $g(\dot{\Gamma})$  and  $C(\phi)$  determine the scaling variable  $\tilde{x} = f(\sigma)g(\dot{\Gamma})C(\phi)/(\phi_0 - \phi)$ , that collapses the viscosity data across all stresses, OSP flow rates, and volume fractions onto a single curve. Importantly scaling variable captures the material dependent response of the suspension and does not affect the universal scaling function or the associated exponents.

To determine the critical exponents governing shear thickening in the presence of OSP flows, we conduct a Cardy scaling analysis [8, 26], and plot the scaling function

$$\zeta^2 \eta = \tilde{x}^2 \mathcal{F}(\tilde{x}) = \mathcal{H} \left( \frac{1}{x_c} - \frac{1}{\tilde{x}} \right) \quad (4)$$

in Fig. 1(d). We find excellent scaling collapse over four orders of magnitude in the scaling variable and seven orders of magnitude in the universal scaling function,  $\mathcal{H}$ . At small values of  $\tilde{x}$ , the data scales with an exponent of -2, as expected for frictionless jamming. As  $\tilde{x} \rightarrow x_c$ , the data show a clear crossover to a different scaling exponent associated with frictional shear jamming. Due to the stress limitations on the ARES-G2 rheometer, we are unable to determine this exponent exactly. Nonetheless, it is consistent with  $\sim 3/2$  reported in previous work [8] (see SI for more details).

Importantly, the scaling function obtained here is the same as that obtained previously [8] (pink line in

Fig. 1(d)). The thickening data for silica and cornstarch suspension from [8] along with the entire range of de-thickening data collapsing onto the same curve, provides further evidence that this function is universal. The collapse of the data onto a universal curve now enables us to determine the exact combination of parameters required to generate an arbitrary viscosity at any applied stress. Indeed the entire system behaviour, across various stresses, volume fractions, and orthogonal flows can be precisely determined by a single material-dependent function,  $C(\phi)$  and two material dependent constants  $\sigma^*$  and  $\Gamma_0$ . Moreover, the universality of the scaling function suggests that these results are generalizable to other geometries and flow types. As such, these results establish a foundation for the precise control and tailoring of the flow behaviour of dense suspensions for applications ranging from the flow of concrete to the conching of chocolate.

Using both the universal and the material specific parameters in the scaling collapse, we generate the evolving shear jamming boundary for this system. The shear jamming surface,  $\phi_J(\sigma, \dot{\Gamma})$ , is governed by the divergence in the scaling function  $\mathcal{F}(\tilde{x})$  at  $\tilde{x} = x_c$ . From Eq. 3, we can write

$$\tilde{x} = \frac{f(\sigma)g(\dot{\Gamma})C(\phi_J)}{(\phi_0 - \phi_J)} = x_c \quad (5)$$

Using  $f(\sigma) = e^{-(\sigma^*/\sigma)^{0.75}}$ ,  $g(\dot{\Gamma}) = e^{-\dot{\Gamma}/\dot{\Gamma}^*}$ , and approximating  $C(\phi)$  as a linear function of  $\phi$  for large volume fractions, we can determine the jamming volume fraction as a function of stress for different values of  $\dot{\Gamma}$  (Fig. 3). Without OSP flows, the shear jamming volume fraction at large stresses is much smaller than that for isotropic frictionless jamming (leftmost line in Fig. 3), consistent with previous literature [7, 27, 28]. At stresses and volume fractions to the left of the jamming line, the suspen-

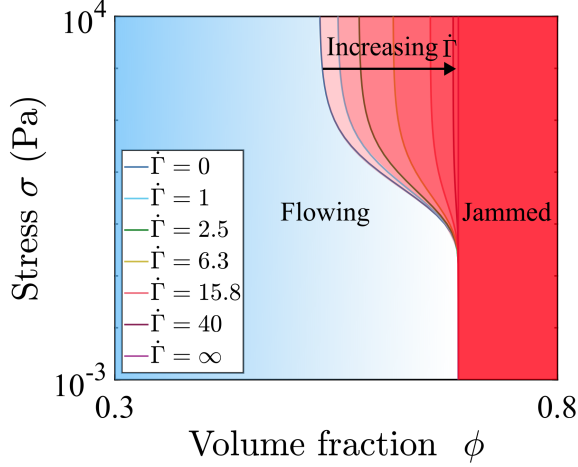


FIG. 3. **Jamming phase diagram as derived from the scaling analysis.** The shear jamming lines for different normalized orthogonal strain rates  $\dot{\Gamma}$ , indicated by the different colours, are drawn in the  $(\phi, \sigma)$  plane. The larger the normalized orthogonal strain rate,  $\dot{\Gamma}$ , the larger the region in which the suspension flows. At small values of  $\dot{\Gamma}$  the suspension is unaffected by orthogonal shear. At large  $\dot{\Gamma}$  however, the formation of force chains is completely impeded, and the volume at which the suspension jams at high stress is equal to the isotropic jamming volume fraction at zero stress.

sion flows and shear thickens, while it becomes jammed on the right side of the line. Typically, this would be the only flow state diagram for any material. Now, however, we can change the location of the jamming line by applying OSP flows. As the OSP strain rate increases, the shear jamming line straightens, with the volume fraction for shear jamming approaching that for frictionless isotropic jamming as  $\dot{\Gamma} \rightarrow \infty$ . Importantly, as this curve is determined by the divergence associated with shear jamming, (i.e. the divergence in the scaling function  $\mathcal{F} \sim (\tilde{x} - x_c)^{-\delta}$ ), it retains the critical exponent associated with frictional shear jamming at all non-zero applied stresses.

Similarly, we find that the discontinuous shear thickening region where  $d \log \eta / d \log \sigma > 1$  shrinks with increasing OSP strain. From Eq. 2, we can write

$$\frac{\sigma}{\mathcal{F}} \frac{d\mathcal{F}}{d\tilde{x}} \frac{g(\dot{\Gamma})C(\phi)}{(\phi_0 - \phi)} \frac{df}{d\sigma} > 1 \quad (6)$$

In the large  $\tilde{x}$  limit where  $\mathcal{F} \sim (x_c - \tilde{x})^\delta$ , this condition reduces to:

$$\frac{-\sigma\delta}{(x_c - \tilde{x})} \frac{df(\sigma)}{d\sigma} \frac{C(\phi)g(\dot{\Gamma})}{(\phi_0 - \phi)} > 1. \quad (7)$$

As  $\dot{\Gamma} \rightarrow \infty$ , i.e. as  $g(\dot{\Gamma}) \rightarrow 0$ , this relationship only holds when  $\tilde{x} \rightarrow x_c$  or equivalently when  $\phi \rightarrow \phi_0$ . Thus, as the

OSP strain rate is increased the boundary for the discontinuous shear thickening region approaches the shear jamming line, shrinking the DST region (see SI for the phase diagram with the DST lines).

The phase diagram in Fig. 3 illustrates how orthogonal shear flows can dramatically alter the suspension properties by driving the system from thickening behaviour governed by the frictional shear jamming critical point towards behaviour governed purely by the frictionless isotropic critical point. The scaling collapse of the data helps uncover the dependence of the viscosity on the OSP flow, with the function  $g(\dot{\Gamma}) = e^{-\dot{\Gamma}/\dot{\Gamma}^*}$  modulating the decrease in the viscosity. As such, OSP flows play a role reciprocal to that played by the shear stress, which increases frictional contacts and drives the system away from frictionless isotropic jamming critical point. This reciprocal behaviour and the striking similarity between the functional forms for  $g(\dot{\Gamma})$  and  $f(\sigma)$ , suggests that their product,  $f(\sigma)g(\dot{\Gamma})$ , dictates the fraction of frictional contacts in the system. This product term captures the effect of the orthogonal perturbations in altering the suspension microstructure, and is analogous to those introduced in previous studies for additional constraints [25] or a minimum strain [13]. This coupling between the primary and the orthogonal flows is more nuanced than the simple additive effect introduced by Brownian motion [29], suggesting that the orthogonal perturbations play a more complex role than simply increasing the effective repulsion between the particles. Nevertheless, we now have an additional tuning knob to alter the suspension behaviour – even at large stresses where the suspension under normal shear will jam, we are able to tune the effective frictional interactions between the particles by imposing orthogonal shear and make the suspension flow. Impressively, we now have complete control over the shear thickening and jamming phase diagram, and can precisely determine the combination of OSP and primary shear rates required to achieve a desired system behaviour. Whether additional strategies for manipulating the suspension, such as acoustics or other time dependent flows, can be treated similarly and folded into this scaling analysis remains an exciting avenue for future research.

Importantly, this new understanding, revealed through the scaling analysis, enables unprecedented control over accessing various regions in the phase diagram. For instance, by switching the orthogonal shear flows on we can drive the system into a high stress flowing state (pink region in Fig. 3). Once these OSP flows are turned off, the system is forced into a previously inaccessible state deep in the shear jammed region. A similar approach can be used to prepare the system deep within the unstable discontinuous shear thickening region. In addition, we can envision using this control over the phase diagram to manipulate the suspension microstructure or embedding memories within these suspensions. As such, this framework opens novel avenues for generating unique mate-

rial states, as well as novel suspension manipulation and training protocols.

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### AUTHOR CONTRIBUTIONS

MR is the corresponding author. BC and IC contributed equally to this work. All the authors were involved in discussions that shaped the idea and experiments in this manuscript. MR ran the experiments. MR, BC, IC, and JPS analyzed the experimental data. MR, BC, JPS and, IC wrote the manuscript with all authors contributing.

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[24] The expert reader may be aware of different exponents reported in the granular literature. We believe these are

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