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Breakup of small aggregates driven by turbulent hydrodynamical stress

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The breakup of small solid aggregates in homogeneous and isotropic turbulence is studied theoretically and by using direct numerical simulations at high Reynolds number, $\mathrm{Re}_\lambda \simeq 400$. We show that turbulent fluctuations of the hydrodynamic stress along the aggregate trajectory play a key role in determining the aggregate mass distribution function. The differences between turbulent and laminar flows are discussed. A definition of the fragmentation rate is proposed in terms of the typical frequency at which the hydrodynamic stress becomes sufficiently high to cause breakup along each Lagrangian path. We also define an Eulerian proxy of the real fragmentation rate, based on the joint statistics of the stress and its time derivative, which should be easier to measure in any experimental setup. Both our Eulerian and Lagrangian formulations define a clear procedure for the computation of the mass distribution function due to fragmentation. Contrary, previous estimates based only on single point statistics of the hydrodynamic stress exhibit some deficiencies. These are discussed by investigating the evolution of an ensemble of aggregates undergoing breakup and aggregation.

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Turbulence has a distinct influence on the aggregation of colloidal and aerosol particles. It not only enhances the rate of collision among particles, i.e., by inducing high velocity differences and preferential concentration within the particle field [1,2], but also it creates hydrodynamic stress that can cause restructuring and breakup of aggregates [3], a phenomenon macroscopically expressed in shear thinning in dense suspensions [4]. The breakup of small aggregates due to hydrodynamic stress in turbulence is of high relevance to various applications, e.g., processing of industrial colloids, nanomaterials, waste waters, and sedimentation of marine snow [3,5]. In a mean-field situation, aggregation-breakup dynamics is described by the Smoluchowski equation. Defining $n_\xi(t) = N_\xi(t)/N_0$, where $N_\xi(t)$ is the number concentration of aggregates consisting of ξ primary particles, and $N_0 = \int_0^\infty d\xi\, \xi N_\xi(t)$, the Smoluchowski equation reads as

$$\dot{n}_\xi(t) = -f_\xi n_\xi(t) + \int_\xi^\infty d\xi'\, g_{\xi,\xi'} f_{\xi'} n_{\xi'}(t) + \frac{3\phi}{4\pi}\left[\frac{1}{2}\int_0^\xi d\xi'\, k_{\xi',\xi-\xi'} n_{\xi'}(t) n_{\xi-\xi'}(t) - n_\xi(t)\int_0^\infty d\xi'\, k_{\xi,\xi'} n_{\xi'}(t)\right], \quad (1)$$

where $\phi = \frac{4}{3}\pi a_p^3 N_0$ is the solid volume fraction, a_p is the radius of the primary particle (assumed monodisperse and spherical), and $k_{\xi,\xi'}$ is the aggregation rate. Breakup is accounted for by the fragmentation rate f_ξ and the fragment distribution $g_{\xi,\xi'}$. Determining these functions is not easy, and despite considerable efforts [6] a basic understanding of breakup dynamics is still lacking. A reason for this is the complex role of turbulence and the way it generates fluctuating stress to which an aggregate is exposed.

The main issue we investigate in this Rapid Communication is how to define and measure the fragmentation rate f_ξ in a turbulent flow. The outcomes of our analysis are manifold: (i) A Lagrangian and an equivalent Eulerian definition of the fragmentation rate can be derived, that fall off to zero in the limit of small aggregate mass, while they have a power-law behavior for large masses; (ii) the power-law tail description is crucial to obtain a steady-state aggregate mass distribution when considering the full aggregation and breakup dynamics; and (iii) turbulent fluctuations allow for a broad asymptotic mass distribution, while a much narrower distribution of aggregates is obtained in the laminar case.

We adopt the simplest possible framework [6], and consider a dilute suspension of aggregates in a stationary homogeneous and isotropic turbulent flow. We consider very small aggregates—much smaller than the Kolmogorov scale of the flow, in the range of 25–100 μm for typical turbulent flows, with negligible inertia. Further, we assume that the concentration of aggregates is such that they do not modify the flow. Hence, their evolution is identical to that of passive pointlike particles (unless extreme deviations from a spherical shape are present, e.g., elongated fibers). Moreover, the aggregates are brittle and breakup occurs instantaneously once they are subject to a hydrodynamic stress that exceeds a critical value [7]. For small and inertialess aggregates, the hydrodynamic stress exerted by the flow is $\sim\mu(\varepsilon/\nu)^{1/2}$, where μ and ν are the dynamic and kinematic viscosity, respectively, and ε is the local energy dissipation per unit mass. Thus, the key role is played by the turbulent velocity gradients across the aggregate which are known to possess strongly non-Gaussian, intermittent statistics [8].

Let $\varepsilon_{\mathrm{cr}}(\xi)$ be the critical energy dissipation needed to break an aggregate of *mass* ξ. In the simplest case of a laminar flow, $\varepsilon_{\mathrm{cr}}$ relates to the critical shear rate for breakup as $G_{\mathrm{cr}} \sim (\varepsilon_{\mathrm{cr}}/\nu)^{1/2}$. Earlier works [7,9] support the existence of a constituent power-law relation for $\varepsilon_{\mathrm{cr}}$, implying that larger aggregates break at a lower stress than smaller ones: $\varepsilon_{\mathrm{cr}}(\xi) = \langle\varepsilon\rangle(\xi/\xi_s)^{-1/q}$, where the exponent q is related to the

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aggregate structure and ξ_s is the characteristic aggregate mass. An equivalent relation exists for small droplets breaking at a critical capillary number $\varepsilon_{\rm cr} \sim \sigma^2/(\mu\rho\xi^{2/3})$ (here ξ is the droplet volume, and σ the interfacial energy). Hence, f_ξ can be equally formulated in terms of a critical dissipation $f_{\varepsilon_{\rm cr}(\xi)}$.

In this contribution we propose to define the fragmentation rate f_ξ or $f_{\varepsilon_{\rm cr}}$ in terms of a *first exit-time* statistics. This amounts to the measurement of the fragmentation rate by using the distribution of the time necessary to observe the first occurrence of a local hydrodynamic stress that is strong enough to break the aggregate. An operational formula of $f_{\varepsilon_{\rm cr}}$ reads as follows: (i) Seed homogeneously a turbulent, stationary flow with a given number of aggregates of mass ξ; (ii) neglect those aggregates in regions where the hydrodynamic stress is too high ($\varepsilon > \varepsilon_{\rm cr}$); and (iii) from an initial time t_0, selected at random, follow the trajectory of each remaining aggregate until it breaks, and count the total number of breaking events in a given time interval $[\tau, \tau + d\tau]$. The time τ is the first *exit time*: For an aggregate initially in a region with $\varepsilon < \varepsilon_{\rm cr}$, τ is the time it takes to the hydrodynamic stress seen by the aggregate along its motion to cross the critical value $\varepsilon_{\rm cr}$ at first opportunity (Fig. 1).

The fragmentation rate is the inverse of the *mean* exit time:

$$f_{\varepsilon_{\rm cr}} = \left[\int_0^\infty d\tau\, \tau \mathcal{P}_{\varepsilon_{\rm cr}}(\tau)\right]^{-1} = \frac{1}{\langle\tau(\varepsilon_{\rm cr})\rangle_{\rm ex}}, \qquad (2)$$

where $\mathcal{P}_{\varepsilon_{\rm cr}}(\tau)$ is the distribution of the first exit time for a threshold $\varepsilon_{\rm cr}$, and $\langle\cdot\rangle_{\rm ex}$ is an average over $\mathcal{P}_{\varepsilon_{\rm cr}}(\tau)$. The definition in Eq. (2) is certainly correct but difficult to implement experimentally as it is needed to follow aggregate trajectories and record the local energy dissipation, something still at the frontier of present-day experimental facilities [10]. The question thus arising is if we can obtain a proxy for the fragmentation rate which is easier to measure. Given a threshold for the hydrodynamic stress $\varepsilon_{\rm cr}$, one can measure the series $T_1, T_2, \ldots$ of *diving times*, namely, the time lags between two consecutive events of instantaneous stress crossing the threshold $\varepsilon_{\rm cr}$ along the aggregate motion (Fig. 1). In Ref. [11], it was proposed to estimate the fragmentation rate as the inverse of the mean *diving time* $f^E_{\varepsilon_{\rm cr}} = 1/\langle T(\varepsilon_{\rm cr})\rangle$. An important result is that $\langle T(\varepsilon_{\rm cr})\rangle$ can be obtained using the Rice theorem for the mean number of crossing events per unit time of a differentiable stochastic process across a threshold [12]. Hence,

$$f^E_{\varepsilon_{\rm cr}} = \frac{1}{\langle T(\varepsilon_{\rm cr})\rangle} = \frac{\int_0^\infty d\dot\varepsilon\, \dot\varepsilon\, p_2(\varepsilon_{\rm cr}, \dot\varepsilon)}{\int_0^{\varepsilon_{\rm cr}} d\varepsilon\, p(\varepsilon)}. \qquad (3)$$

Here the numerator is the Rice formula giving the mean number of crossings of $\varepsilon_{\rm cr}$ in terms of the joint probability of dissipation and its time derivative $p_2(\varepsilon, \dot\varepsilon)$; the denominator is the measure of the total time spent in the region with $\varepsilon < \varepsilon_{\rm cr}$. Notice that the integration in the numerator goes only on positive values in order to consider only upcrossing of the threshold $\varepsilon_{\rm cr}$ [12]. An obvious advantage of Eq. (3) is that it is *quasi-Eulerian*: It does not require to follow trajectories, since it depends only on the spatial distribution of dissipation and of its first time derivative in the flow. Expressions (2) and (3) are not strictly equivalent. A direct calculation of the mean exit time in terms of the distribution of diving times gives indeed $\langle\tau(\varepsilon_{\rm cr})\rangle_{\rm ex} = \langle T^2(\varepsilon_{\rm cr})\rangle/[2\langle T(\varepsilon_{\rm cr})\rangle]$, which relates the mean exit time to moments of the diving time.

Once a definition from first principles is set up, we proceed to measure the fragmentation rate for aggregates convected as passive point particles in a statistically homogeneous and isotropic turbulent flow, at Reynolds number $\mathrm{Re}_\lambda \simeq 400$. Details on the direct numerical simulations (DNSs) of Navier-Stokes equations with 2048^3 grid points and Lagrangian particles are in Ref. [2]. The present analysis is obtained averaging over 6×10^5 trajectories, recorded every $0.05\tau_\eta$, where τ_η is the Kolmogorov time of the flow.

Figure 2 shows the fragmentation rate measured from the DNS data following the evolution of the velocity gradients along aggregate trajectories. The exit-time measurement (2) and its Eulerian proxy (3) show a remarkable, nontrivial behavior for small values of the critical threshold, i.e., for large aggregate mass. In this region, there is a competing effect between the ease in breaking a large aggregate and

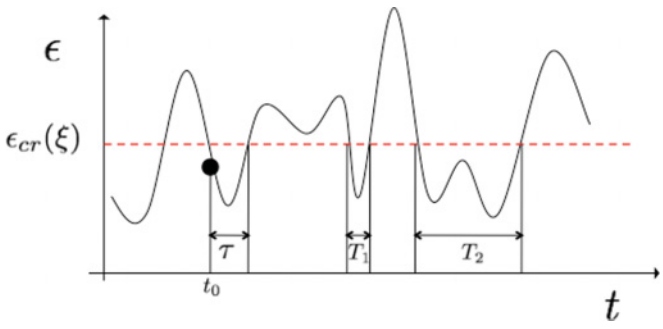

FIG. 1. (Color online) Pictorial evolution of the energy dissipation ε along an aggregate trajectory. Starting to record the stress at time t_0, the exit time τ and the diving times $T_1, T_2, \ldots$ for a given threshold $\varepsilon_{\rm cr}$ are shown.

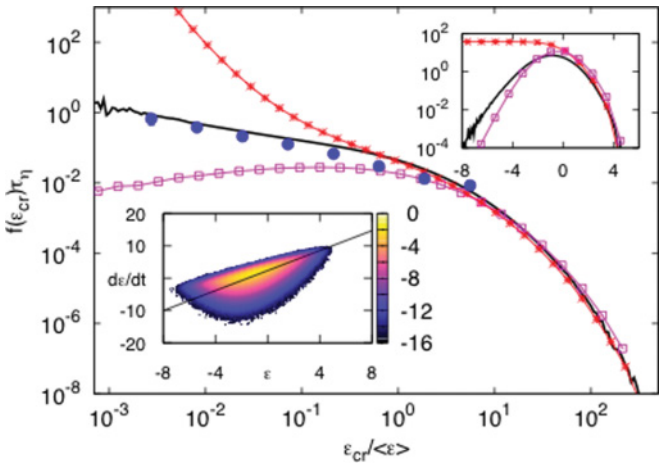

FIG. 2. (Color online) The normalized fragmentation rate $f_{\varepsilon_{\rm cr}}\tau_\eta$ vs the normalized energy dissipation $\varepsilon_{\rm cr}/\langle\varepsilon\rangle$. Solid circles are the definition (2), measured up to thresholds where statistical convergence of exit times is obtained; the solid line is $f^E_{\varepsilon_{\rm cr}}$. Squares are $f^{\rm I}_{\varepsilon_{\rm cr}}$, while crosses are $f^{\rm II}_{\varepsilon_{\rm cr}}$. Bottom inset: Joint distribution $p_2(\varepsilon, \dot\varepsilon)$. The continuous line is the dimensional estimate $\dot\varepsilon \sim \varepsilon/\tau_\eta(\varepsilon)$. Top inset: Numerator of Eqs. (3)–(5); curve colors are the same as in the main figure.

the difficulty in observing a large aggregate existing in a region of low-energy dissipation. As a result, the estimated fragmentation rate develops a quasi-power-law behavior for small thresholds. On the other hand, for large thresholds the superexponential falloff is expected. It is the realm of very small aggregates that are broken only by large energy dissipation bursts. The exit-time (2) and diving-time (3) measurements are very close and we therefore consider the latter a very good proxy of the former, the real fragmentation rate. The main advantage of the estimate (3) is the very high statistical confidence that can be obtained since it is a quasi-Eulerian quantity. Moreover, it gives a reliable estimate also in the region of large thresholds where the convergence of exit-time statistics, requiring very long aggregate trajectories, is difficult to obtain.

Starting from Eq. (3), simple models can be proposed for the statistics of $\dot{\varepsilon}$, whose direct measure requires a very high sampling frequency along Lagrangian paths. First, as it appears from the bottom inset of Fig. 2, the dissipation and its time derivative are significantly correlated. Scaling on dimensional grounds suggests $\dot{\varepsilon} \sim \varepsilon/\tau_\eta(\varepsilon)$, where $\tau_\eta(\varepsilon) \sim (\nu/\varepsilon)^{1/2}$ is the local Kolmogorov time. It follows that the joint probability density function (PDF) $p_2(\varepsilon,\dot{\varepsilon})$ can be estimated as $p_2(\varepsilon,\dot{\varepsilon}) = \frac{1}{2}p(\varepsilon)\delta(|\dot{\varepsilon}| - \varepsilon/\tau_\eta(\varepsilon))$, where $p(\varepsilon)$ is the probability density of energy dissipation. The prefactor $1/2$ appears since for a stationary process $\dot{\varepsilon}$ is positive or negative with equal probability. Plugging this expression in Eq. (3) gives

$$f^{\mathrm{I}}_{\varepsilon_{\mathrm{cr}}} = \frac{\frac{1}{2}\varepsilon_{\mathrm{cr}}\, p(\varepsilon_{\mathrm{cr}})/\tau_\eta(\varepsilon_{\mathrm{cr}})}{\int_0^{\varepsilon_{\mathrm{cr}}} d\varepsilon\; p(\varepsilon)}. \tag{4}$$

We refer to it as *closure I*. A different approach was proposed in Ref. [6]. It assumes that active regions in the flow where $\varepsilon > \varepsilon_{\mathrm{cr}}$ engulf the aggregates at a rate $\sim 1/\tau_\eta(\varepsilon)$, which results in

$$f^{\mathrm{II}}_{\varepsilon_{\mathrm{cr}}} = \frac{\int_{\varepsilon_{\mathrm{cr}}}^{\infty} d\varepsilon\; p(\varepsilon)/\tau_\eta(\varepsilon)}{\int_0^{\varepsilon_{\mathrm{cr}}} d\varepsilon\; p(\varepsilon)}. \tag{5}$$

We refer to it as *closure II*. Both models share the advantage of being fully Eulerian and based on the spatial distribution of the energy dissipation only. In Fig. 2, the fragmentation rates obtained from closures I and II are also shown. Both of them reproduce the correct behavior for large values of the critical dissipation but deviate for small ones. The reason for such a discrepancy is made clear in the top inset of Fig. 2, where the numerator of Eqs. (4) and (5) is shown. It appears that, for small values of the critical stress $\varepsilon_{\mathrm{cr}}$, $f^{\mathrm{I}}_{\varepsilon_{\mathrm{cr}}}$ underestimates the number of breakup events, while the numerator of $f^{\mathrm{II}}_{\varepsilon_{\mathrm{cr}}}$ saturates to a constant value.

Since in experiments breakup *a fortiori* takes place together with aggregate recombination, we explore how the actual fragmentation rate Eq. (3) and the two closures, Eqs. (4) and (5), influence the time evolution of an ensemble of aggregates $n_\xi(t)$. At this purpose, the Smoluchowski equation (1), subject to the initial condition $n_\xi(0) = \delta(\xi - 1)$, is evolved in time. To model aggregation we use the classical Saffman-Turner expression $k_{\xi,\xi'} = D_0/\tau_\eta(\xi^{1/d_f} + \xi'^{1/d_f})^3$, where D_0 is an $O(1)$ constant and d_f is the fractal dimension that relates the collision radius of an aggregate to its mass, $a/a_p = \xi^{1/d_f}$. Breakup is assumed to be binary and symmetric, $g_{\xi,\xi'} = 2\delta(\xi - \xi'/2)$, which, despite its simplicity, represents well the quality of the evolution. To quantify our findings we consider the second moment observable readily accessible from static light scattering [7]. Figure 3 shows the time evolution of I_0 for typical values of d_f and q found in turbulent aggregation of colloids [3]. After an initial growth period, curves obtained with Eqs. (3) and (5) both relax to a steady state. At small solid volume fraction ϕ, the two models nearly overlap, whereas at larger ϕ closure II underestimates I_0. This behavior is confirmed in the inset of Fig. 3, which shows I_0 at steady state for both models and for different values of ϕ. Clearly, at

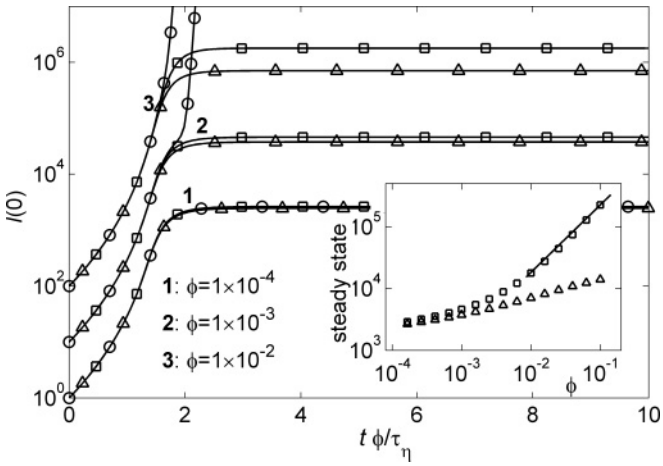

FIG. 3. Time evolution of I_0 for $d_f = 2.4$ and $q = 0.36$ with breakup rates $f^{E}_{\varepsilon_{\mathrm{cr}}}$ (squares), $f^{\mathrm{I}}_{\varepsilon_{\mathrm{cr}}}$ (circles), and $f^{\mathrm{II}}_{\varepsilon_{\mathrm{cr}}}$ (triangles). Runs with different solid volume fractions ϕ are shifted. Inset: I_0 at steady state as a function of ϕ (same symbols). At large ϕ, the model with fragmentation (3) relaxes to the predicted scaling curve $I_0 \sim \phi^{1/(1+\chi/q-3/d_f)}$, given by the solid line. Here, $\xi_s = 10^4$, implying that $a/a_p \sim \xi_s^{1/d_f} \approx 50$ is the value of the characteristic aggregate size.

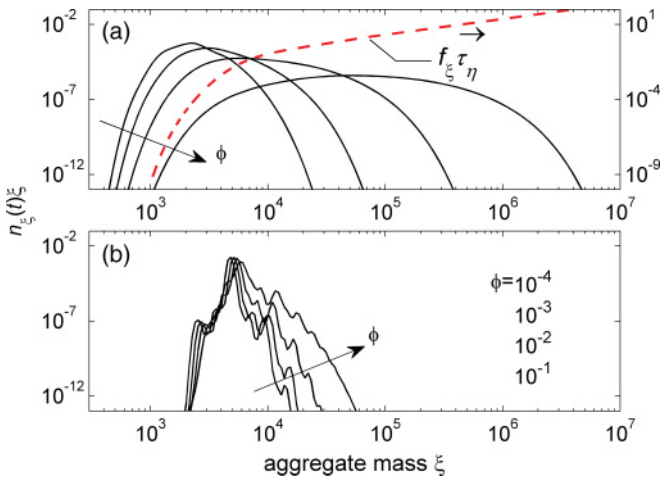

FIG. 4. (Color online) Stationary mass distribution $n_\xi(t)$ with $d_f = 2.4$ and $q = 0.36$, for different solid volume fractions ϕ. Distributions obtained in the turbulent flow at $\mathrm{Re}_\lambda \simeq 400$ (a) are compared to those of the laminar cases (b), with the uniform shear rate $\varepsilon^{\mathrm{lam}}_{\mathrm{cr}} = \langle\epsilon\rangle$. In (a), we plot f_ξ (dashed line) as given by Eq. (3), assuming a power-law behavior in the limit of large aggregate mass.

large ϕ other phenomena, i.e., modulation of the flow due to the particles, may occur which, however, is beyond the scope of the present work. Closure I shows a very different behavior. At small ϕ, an evolution similar to previous cases is observed. However, by increasing ϕ a drastic change appears and I_0 diverges, a direct consequence of the presence of a maximum in the shape of $f^{\mathrm{I}}_{\varepsilon_{\mathrm{cr}}}$ (see Fig. 2).

The time evolution of the number concentration $n_\xi(t)$, governed by Eq. (3), is further examined in Fig. 4. Here, we compare the steady distribution obtained in the turbulent flow with that of a laminar flow for different values of the solid volume fraction. In the turbulent case, $n_\xi(t)$ rapidly grows and reaches a stationary state whose peak mode is controlled by the magnitude of aggregation, i.e., the solid volume fraction ϕ. Increasing ϕ causes the mode to broaden and to shift to the right as the aggregation gets more pronounced. The distribution thus gradually moves into the region where $f_{\varepsilon_{\mathrm{cr}}}$ assumes power-law behavior. A numerical fit of the left fragmentation tail in Fig. 2 gives $f_\xi \sim \xi^{\chi/q}$, with $\chi = 0.42 \pm 0.02$ (dashed curve in Fig. 4). Using this latter expression in Eq. (1), one can derive a scaling relation for integral quantities of $n_\xi(t)$ at steady state [13,14]. Such scaling is reported in the inset of Fig. 3. On the other hand, in the laminar case where a uniform shear rate governs the breakup, the steady-state distribution is much narrower and shows multiple resonant modes. These are due to the sharp onset of breakup once the aggregates grow larger than the characteristic aggregate mass ξ_s. These results clearly demonstrate the strong influence of turbulent fluctuations on the statistically stationary mass distribution function.

We have presented a study of the fragmentation rate of small and diluted aggregates in turbulent flows at high Reynolds number. We have introduced an expression for the fragmentation rate in terms of the *exit-time* statistics, which is a natural way of measuring the first-order rate process. Also, a purely Eulerian proxy based on Eq. (3) provides a very good approximation to the actual fragmentation rate measured from our DNS. Remarkably, a steady state in the full breakup-aggregation process is crucially determined by the left tail of the fragmentation rate, i.e., by events of low-energy dissipation. Our investigation puts the basis for many developments, such as the stability of the Smoluchowski evolution using the measured fragmentation rates in experiments, and the extension to the case of *inertial* aggregates. In such case, the correlation between the hydrodynamic shear and the Stokes drag may result in a nontrivial breakup rate dependency on the degree of inertia. Future work aims to introduce spatial fluctuations in the mass distribution caused by local breakup, a research path still poorly explored.

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