Nonlinear Bubble Dynamics in a Slowly Driven Foam

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Sudden topological rearrangement of neighboring bubbles in a foam occur during coarsening, and can also be induced by applied forces. Diffusing-wave spectroscopy measurements are presented of such dynamics before, during, and after an imposed shear strain. The rate of rearrangements is proportional to the strain rate, and the shape of the correlation functions shows that they are spatially and temporally uncorrelated. Macroscopic deformation is thus accomplished by a nonlinear microscopic process reminiscent of dynamics in the propagation of earthquake faults or the flow of granular media.

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Aqueous foams consist of a dense random packing of gas bubbles stabilized by surface active macromolecules [1,2]. The bubble shapes can vary from nearly spherical to nearly polyhedral, forming a complex geometrical structure insensitive to details of the liquid composition or the average bubble size. As a form of matter, foams exhibit remarkable mechanical properties that arise from this structure in ways that are not well understood. Namely, foams can support static shear stress, like a solid, but can also flow and deform arbitrarily, like a liquid, if the applied stress is sufficiently large [3,4]. The solidlike properties are due to surface tension and the shape distortion of bubbles in linear response to a small applied strain. The liquidlike properties, however, cannot be similarly understood by linear response since large deformations, though macroscopically homogeneous, are accomplished by microscopically inhomogeneous neighbor-switching rearrangements of bubbles from one tightly packed configuration to another. Intermittent structural rearrangements also occur in quiescent foams due to the alteration of packing conditions from the diffusion of gas from smaller to larger bubbles [5–8]. No matter what the driving force, all such dynamics are highly nonlinear and complex, involving abrupt topology changes and large local motions that depend on structure at the bubble scale. For example, in the Princen-Prud'homme model of foam as a two-dimensional periodic array of hexagonal bubbles, topological rearrangements happen instantaneously and simultaneously throughout the entire sample [9,10]. In a more realistic dense random packing of bubbles, however, the rearrangement events can be *localized,* occurring with variable size and duration in different regions at different times. The influence of randomness on the link between microscopic structure and macroscopic deformation has been studied by computer simulation [5,11–14]. Recently, Okuzono and Kawasaki [15] predicted that rearrangements in a slowly driven foam have a broad, power-law, distribution of event rate vs energy release, and thus exhibit self-organized criticality.

Experimentally, rearrangement phenomena are difficult to study because the opaque nature of foams restricts direct visualization to bubbles near a surface. Here, we report multiple light scattering measurements that take advantage of this property to probe the microscopic, bubble-scale, response of a bulk foam to a macroscopically imposed shear strain. The average bubble size is monitored from the average transmitted intensity, while the nature and rate of rearrangements are monitored from the intensity fluctuations via diffusing-wave spectroscopy (DWS) [16]. In addition to studying the link between macroscopically homogeneous shear strain and microscopically inhomogeneous bubble rearrangements, we also investigate the interrelationships between shear-induced dynamics and stability. Since foams are nonequilibrium systems, it is crucial to understand how foam rheology and bubble dynamics are affected by evolution, and, conversely, how foam stability is affected by flow.

Our measurements are performed on a commercial shaving foam which has been previously characterized by DWS [6,17]. It consists of polydisperse gas bubbles, 92% by volume, and coarsens by the diffusion of gas from smaller to larger bubbles; drainage and film rupture are not significant. The foam structure and its evolution are monitored from the probability T for incident light to be transmitted through an opaque slab of thickness L using the diffusion theory prediction, $T = (1 + z_e)/(L/l^* + 2z_e)$ where $z_e = 0.88$ and l^* is the photon transport mean free path [18]. For our material, l^* is 3.5 times the average bubble diameter, and transmission measurements show that it grows as nearly the $\frac{1}{2}$ power of time after 20 min. Such growth indicates a highly reproducible, self-similar bubble size distribution that is independent of initial conditions. Most measurements reported here were therefore taken after the foam had aged about 100 min, well into the scaling regime, where the average bubble diameter is 60 μm.

In order to shear the foam while simultaneously probing its bubble-scale response with DWS, we confine samples between parallel glass plates, 13×46 cm^2, arranged such that the narrow ends are open and that one plate can be slid at constant velocity over the other. To ensure that the strain is homogeneous and that the light scattering measurements and analyses are sound, we vary the plate spacing to be $L = 6$, 8, or 10 mm, and apply shear by

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motion of the plate on either the incident or transmission sides. To minimize wall slip, both plates are rendered hydrophobic by treatment with dichlorodimethylsilane, and different combinations of smooth and roughened (via sandblasting) surfaces are employed. A previous study of strain-induced dynamics used flow through a constriction, where the strain is inhomogeneous and even involves plug glow [19,20].

The bubble rearrangement dynamics induced by either coarsening or application of shear were probed by fluctuations in the intensity of a speckle of transmitted light at $\lambda = 488$ nm. Results are expressed by the normalized electric field autocorrelation function, $g_1(\tau)$, vs the delay time τ. The inset of Fig. 1 shows typical data for 2 min collection durations before, during, and after shear was applied at rate 0.5 s^{-1}. In all three cases, the shape of $g_1(\tau)$ is nearly a single exponential as described by the thick-sample prediction of the theory of DWS for uncorrelated dynamics, $\sqrt{6\Gamma_1\tau}/\sinh\sqrt{6\Gamma_1\tau}$, where the first cumulant, Γ_1, or initial decay rate, is adjusted to fit the data. The quality of such fits, while always satisfactory, is best for quiescent samples and small strain rates. Introducing a second parameter for correlated motion due to a velocity gradient, as in Refs. [21,22], improves the fits but does not affect the result for Γ_1, showing that it provides a robust characterization of $g_1(\tau)$. Its time evolution is shown in the main plot of Fig. 1, and is contrasted with results for a quiescent, unsheared, foam. Evidently Γ_1 is larger during shear and smaller afterwards, as seen in previous work [19], but recovers to the quiescent behavior within about 10 min, demonstrating that any change caused by shear heals away in a coarsening time.

Results from monitoring the foam structure via the transmission probability, T, simultaneously with $g_1(\tau)$, show that application of shear affects the dynamics but not the average bubble size. The evolution of Γ_1 and l^* for a foam sheared at a steady rate of 0.5 s^{-1} until 130 min of age, by which time the average bubble diameter has grown by a factor of 3, is contrasted in Fig. 2 with results for a quiescent foam. The results for Γ_1 are larger during shear, and recover to the quiescent behavior soon after the shear is stopped. However, the results for l^*, and therefore the average bubble size, are identical for sheared and quiescent samples. Thus, in contrast with a recent prediction [13], *shear has minimal affect on foam structure and none at all on the coarsening processes.*

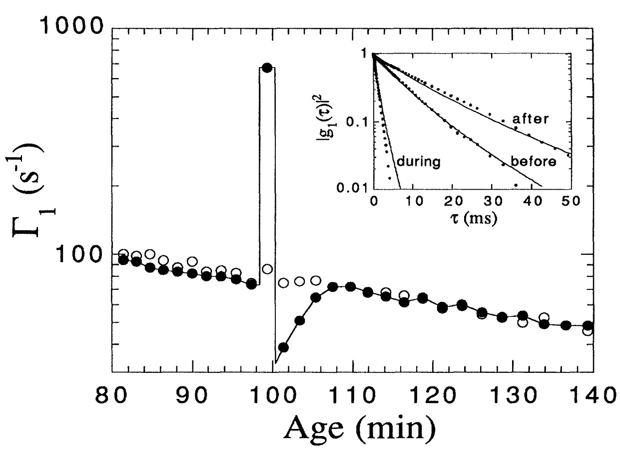

FIG. 1. First cumulant vs foam age for a quiescent sample (open circles) and one that is sheared from 98 to 100 min (solid circles connected by lines); the plate velocity for the latter is $V = 4$ mm/s, and the plate separation is $L = 8$ mm. The inset shows raw autocorrelation data (symbols) and fits (curves) before, during, and after application of shear.

The effect of shear is primarily to rearrange bubbles without causing change in average size or rate of coarsening. Nevertheless, Γ_1 is temporarily suppressed following the cessation of shear. The magnitude and duration of the effect can be understood as follows. Since coarsening-induced rearrangements occur at random, there must be a random spatial distribution of regions under different local strains. Some regions have recently undergone rearrangement and are therefore under low strain, while in others the bubbles are distorted away from their relaxed shapes and are on the threshold of rearrangement. To the extent that flow homogenizes the strain field, the rate of coarsening-induced rearrangements should be suppressed and should remain so until inhomogeneities are reestablished during coarsening. This would require on the order of 10 min for 100 min old foam samples, consistent with all our recovery data independent of the size of the suppression. Furthermore, the number of rearrangements induced by sudden shear, and the corresponding suppression in dynamics afterwards, should depend on the total applied strain. This is tested in Fig. 3, where the fractional suppression in the cumulant following shear is plotted as a function of applied strain. These data were obtained with the roughened glass plates, and the strain rate was always larger than 0.5 s^{-1}. Data for the two sample thicknesses are indistinguishable, and show the expected behavior. First, there is no effect below a strain of about 5%, which may therefore be identified as the yield strain below which no rearrangements

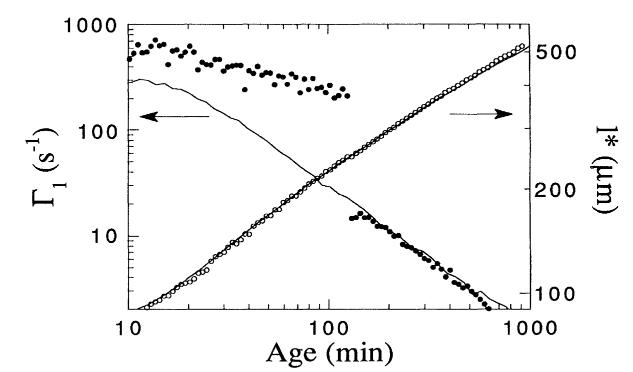

FIG. 2. First cumulant (closed circles, left axis) and transport mean free path (open circles, right axis) vs age for foam sheared at rate 0.5 s^{-1} until 130 min; results for an unsheared sample are shown for comparison by solid curves. Note that flow affects Γ_1 but not l^*.

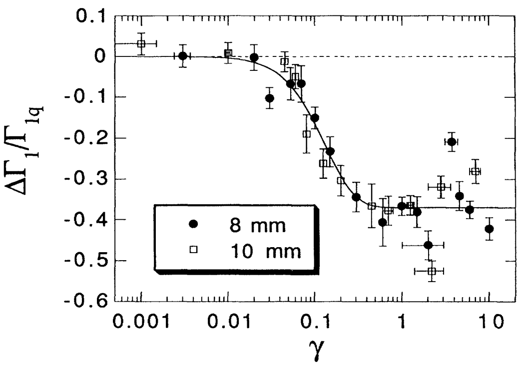

FIG. 3. Fractional change in the cumulant immediately following a shear step strain of size γ; the plate separations are as labeled, and the solid curve is a guide to the eye. Results are for roughened plates, and uncertainty in strain due to both uncertainty in total plate motion and wall slip is denoted by horizontal error bars.

are induced. Second, the observed suppression saturates above a strain of about 1, which may therefore be identified as the strain required to induce rearrangements everywhere and thus destroy correlations caused by coarsening. It is not understood why the degree of suppression is limited to only about 45%, but may reflect the difficulty of achieving an optimal random close-packed configuration with minimal strain inhomogeneity. Nevertheless, it is clear that the influence of shear on our foam is limited to inducing rearrangements and thereby destroying subtle correlations in the strain field that arise from the evolution process.

The nature and the rate of the rearrangement dynamics caused by shear can now be studied quantitatively through $g_1(\tau)$. If the motion of scattering sites in a diffuse photon path are random and uncorrelated, the theory of DWS predicts $g_1(\tau)$ to have the observed nearly exponential form with $\Gamma_1 \cong (L/l^*)^2/\tau_0$, where τ_0 is the average time for a single scattering site to move by a distance $k^{-1} = \lambda/2\pi$ [16]. This has been previously observed for both Brownian motion, where $\tau_0 = 1/Dk^2$ is set by the particle diffusion coefficient, and the sudden rearrangement of neighboring bubbles induced by coarsening, where τ_0 is proportional to the average time between rearrangements at a single scattering site [6]. For a quiescent sample at 100 min, the value of this characteristic time scale is $\tau_{0q} = 20$ s [17], much shorter than both the 2 min run durations and the 10 min needed for noticeable coarsening. The essentially exponential decay observed in Fig. 1 for the same foam under shear shows that strain-induced rearrangements are also uncorrelated. By contrast, if the shear deformation were homogeneous down to the bubble scale, then $g_1(\tau)$ would decay exponentially with τ^2 [21,22]. Therefore, macroscopically homogenous shear deformation is accomplished by a series of discrete microscopic bubble rearrangements that occur intermittently throughout the foam.

The average time τ_0 between random, uncorrelated bubble rearrangements can be gauged using the relation $\Gamma_1 \cong (L/l^*)^2/\tau_0$. Since flow has no effect on l^*, the entire change in Γ_1 is due to a change in the rate $1/\tau_0$. Therefore, the ratio of this rate in a sheared foam to that of a quiescent, unsheared, sample with the same thickness is given by $\tau_{0q}/\tau_0 = \Gamma_1/\Gamma_{1q}$, independent of L, and can be deduced for a single sample by measurements of the type in Fig. 1. Results are shown in Fig. 4 as a function of plate velocity for three slab thicknesses and smooth vs rough surface preparations. The two relevant time scales are V/L, the strain rate in the absence of wall slip, and $1/\tau_{0q}$, set by the rate of rearrangements in a quiescent sample; therefore, data are plotted vs the Deborah number $(V/L)\tau_{0q}$. The good collapse of data for different thicknesses, but not for different surface separations, implies that the strain rate is macroscopically uniform across the sample, though it may be less than or equal to V/L. In all cases, Γ_1/Γ_{1q} approaches 1 for small $(V/L)\tau_{0q}$, and increases monotonically for larger values of $(V/L)\tau_{0q}$.

The behavior seen in Fig. 4 can be explained as follows. The rate of *coarsening-induced* rearrangements is always proportional to $1/\tau_{0q}$, while the rate of *shear-induced* rearrangements, on the other hand, is a yield strain divided by the time needed to accumulate that strain via homogenous deformation, and is therefore equal to the applied strain rate. The latter has been observed in computer simulation [14] and suggested in analysis of flow experiments [20,23]. If these two processes are independent, the total rate of rearrangements is the sum of their individual rates, giving $\Gamma_1/\Gamma_{1q} = 1 + A(V/L)\tau_{0q}$ where A is a constant that increases with decreasing wall slip and with increasing rearrangement event size. By adjusting A, an excellent fit is obtained to the data in Fig. 4 for the case of roughened surfaces; the systematic deviation for the case of smooth surfaces could be due, e.g., to a degree of wall slip that increases with

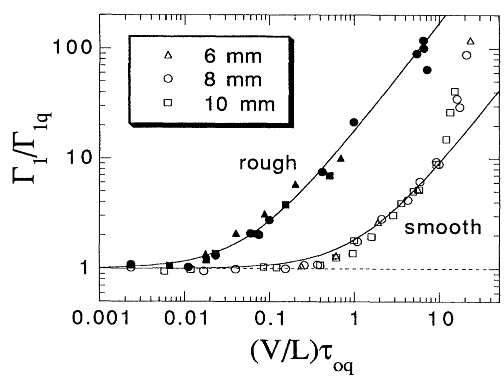

FIG. 4. Ratio of the first cumulant for a foam under shear to that of a quiescent sample of the same thickness, L, vs Deborah number; V is the velocity difference imposed between top and bottom plates and $\tau_{0q} = 20$ s. Open (solid) symbols are for smooth (roughened) plates. Solid curves are fits, indicating that coarsening-induced rearrangements dominate at small $(V/L)\tau_{0q}$ while shear-induced rearrangements increase proportional to the strain rate.

V/L. This supports our picture and also shows that flow at small strain rates is accomplished through time evolution according to the rate of coarsening-induced rearrangements rather than the rate of change of the bubble size distribution. Furthermore, the fitting result $A = 17$ for the case of minimal wall slip is sensible. The rearrangement event size as measured by DWS is the entire region where bubble displacement exceeds λ; this includes a core region where bubbles undergo topological rearrangement as well as a shell of surrounding bubbles that respond elastically. For the case of no wall slip, A must equal the ratio of event volume to core volume since only the latter contributes to the flow. If the event volume is 10 bubbles across, as estimated for coarsening [6], then the inner core undergoing rearrangement is $10/\sqrt[3]{A} \cong 4$.

While the observed dynamics is a nonlinear stick-slip process reminiscent of avalanches in models of granular media and earthquakes, our DWS measurements show that there is a single characteristic time scale, τ_0, between shear-induced events. The simplest explanation is that events have a characteristic volume ν, as discussed above, and are initiated at rate per unit volume R such that $\tau_0 = 1/R\nu$. However, this would contradict the self-organized criticality prediction of Ref. [15], since a broad distribution of event sizes and rates would generally give rise to a broad distribution of times between events and a correspondingly stretched exponential decay of $g_1(\tau)$. These simulations are for a two-dimensional foam with zero liquid content, ignoring coarsening and conservation of film material, but are otherwise realistic. Nevertheless, some such omission may probe unwarranted because it is difficult to reconcile the predictions with our observations. For example, large events may be killed by the finite system size or strain rate, and the cutoff would correspond to the characteristic time observed here. The finite system size is not a possibility, however, since our results are independent of sample thickness. To determine whether the strain rate is finite, we compare with the time scale for completion of rearrangements, which for our material is 0.1 s [23]. The strain rate is therefore infinitesimal for $(V/L)\tau_{0q} \ll 200$, which is well satisfied here. Another possibility is that there could be a spectrum of event sizes such that $R\nu \propto V/L$; however, an intrinsic cutoff would still be required since we see no dependence of the dynamics on L. These scenarios could be explored by further simulation of the mean-squared change in position of the scattering sites, as well as the event rate vs spatial size. Alternative models relaxing the assumptions of Ref. [15] would also be useful.

The study of sudden rearrangements of bubbles in a slowly driven foam offers a unique opportunity for general insight into stick-slip dynamics. By contrast with systems such as granular media or earthquake faults, where microscopic details can only be caricaturized, the microscopic physics of soap films is well understood and the structural randomness of the bubble packing is a natural and highly reproducible feature. Furthermore, the nonlinear rearrangement dynamics in foams can be directly probed by DWS as demonstrated here. Reconciling the contradictions between our observations and computer simulations should lead to a deeper understanding of not only stick-slip dynamics in general, but of the interrelationships between the structure, stability, and rheology of an important class of disordered materials as well.

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