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# A Diffusing Wave Spectroscopy Study of Constrictive Flow of Foam

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PACS.83.70.Hq – Heterogeneous liquids: suspensions, dispersions, emulsions, pastes, slurries, foams, block copolymers, etc.

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**Abstract.** — The dynamics of a model foam in constrictive flow have been studied by diffusing-wave spectroscopy. Below a threshold strain rate the rate at which elementary strain-induced rearrangements occur in the foam depends only on the strain, whereas above this threshold the rate is simply proportional to strain rate. A possible mechanism for the unexpected behaviour at low strain rates is outlined.

## 1. Introduction

Foams are complex and disordered structures of both fundamental and technological interest [1, 2]. From a practical view-point, their mechanical response is amongst their more important properties [3]. At low stress foam behaves as an elastic solid, while above a reasonably well-defined yield stress it deforms plastically. This microscopic behavior must involve local rearrangements of the bubbles comprising the foam. Until very recently such elementary rearrangements have been experimentally unobservable, except for the two-dimensional case. However, the recently developed technique of diffusing wave spectroscopy (DWS) [4, 5] permits the rate of occurrence of such events to be studied in real foam. Two studies of flowing foam by DWS have shown that such rearrangements are promoted by strain in a flowing foam [6], and that, to first approximation, their rate depends linearly upon strain rate [7]. The present paper considers certain unexpected threshold phenomena observed in the constrictive geometry of the foam flow cell used in these studies.

Much of our understanding of foam rheology rests upon computer simulations [8] which have become more realistic in recent years, extending to encompass not only disorder [9–11], but also the inclusion of a liquid fraction in the foam [12]. Plastic deformation is found to occur via a succession of localized elementary events in which a group of bubbles rearranges to relieve local stress. (In wet foams, such as those studied here, basic topological changes such as the  $T_1$  process in two dimensions probably do not occur.) In simulations of ideal dry froth (volume fraction of gas  $\phi = 1.0$ ) the yield stress corresponds to an imposed strain exceeding a value of

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about 0.5 [10]. Continued application of this stress leads to indefinite increase of strain: the foam flows. Reduction of the gas fraction causes the critical value of strain to fall; it may reach zero at a critical  $\phi$  ( $= 0.84$ ) [12]. At a given strain above the critical value, rearrangement occurs whenever the accumulated stress at a particular point in the foam exceeds some critical level. The rate of rearrangements should thus be proportional to the strain rate.

In foam light is highly multiply scattered: it propagates by diffusion [4]. Interference between light following several paths through the foam gives rise to flicker—fluctuations in the detected intensity. The phase change of  $\approx \pi$  necessary to cause a change from constructive to destructive interference is here accumulated in events distributed over the *entire* light path, rather than in a single event as in conventional dynamic light scattering [13]. In studies of foam [6,7,14–16] DWS is used to probe spatially or temporally rare events involved in the coarsening or flow of foam. For static foam, the fundamental process involved in the temporal evolution of the foam is an elementary rearrangement event: a local neighbourhood some ten bubbles across spontaneously reorders as coarsening causes a critical value of the local stress to be exceeded [14, 16].

A brief review of the present understanding of the DWS signal in foam is appropriate [14]. The photon correlation functions observed for flowing foams are basically exponential in form [6], as for static foam [14, 15]. The initial decay constant

$$\Gamma_1 = \left(\frac{L}{l^*}\right)^2 \tau_0^{-1} \quad (1)$$

relates to the probability of a bubble rearrangement event affecting light as it traverses the sample.  $L$  is the sample thickness and  $l^*$  the transport mean free path of the light, the average distance a photon travels before its propagation direction is randomized. It is suggested [14] that  $\tau_0$  reflects the average time interval between rearrangement events at any single point in the foam:  $\tau_0^{-1} \approx Rr^3$  where  $R$  is the average rate of rearrangements per unit volume and  $r$  is the radius of the volume of foam involved directly in a typical event.

For laminar flow of simple fluids the correlation functions measured by DWS show an exponential decay quadratic in time, governed by the velocity gradient parallel to the incident light beam [17]. However, in the present case this velocity gradient is negligible as foam, when slowly flowing in a smooth-walled vessel, displays plug flow [3] (visual observation of the flowing foam in the cell used in the present experiments confirms the plug nature of the flow, the foam moving as a unit apart from a single boundary layer of bubbles [6]). Thus the deterministic flow of the foam does not, in itself, contribute to the time dependence of the observed correlation functions, which relates to random rearrangements as for static foam.

## 2. Experimental

Our methods have been described elsewhere [6, 7] and need only be summarized. The experimental material was Gillette shaving foam [18], a complex aqueous surfactant solution which produces stable and reproducible foam samples, with  $\phi = 0.92 \pm 0.01$  [14].

The central part of our foam flow cell is shown diagrammatically in Figure 1. The entire cell was 55 cm long, comprising two glass plates separated by 5 mm wide aluminium spacers shaped to form a parallel-sided channel narrowing in the centre. The height of the cell ( $l$ ) varied as sketched in the figure. The cell was sealed by two PTFE barriers which could be moved in parallel, forcing the cell contents along the cell and through the central constriction. In use, one barrier was removed, the cell was filled with foam, care being taken to exclude air, and the barrier was replaced to seal the foam into the cell. Barrier motion was induced with the pusher mechanism of an infusion pump, the barrier speeds being  $\lesssim 1$  mm/min (this speed

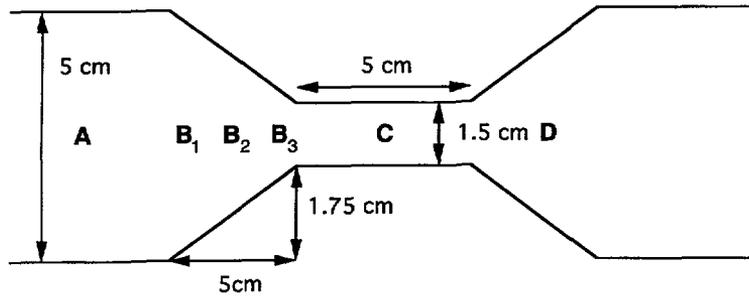


Fig. 1. — A sketch (not to scale) of the central portion of the flow cell used. DWS observations were made at various points in the regions A, B, C and D. See the text for further description.

is referred to below as the 'flow rate'  $V$ ). As foam flows through the cell the strain is constant at a given point in the cell, but the strain rate can be varied by changing the flow rate.

A laser beam was normally incident on the cell and light forward scattered at  $0^\circ$  was detected. From the intensity autocorrelation function of the scattered light the field correlation function,  $g_T^{(1)}(\tau)$ , was extracted. Observations were made at various points on the centre-line of the flow cell (see Fig. 1): two in regions of constant strain (C is at the mid-point of the constriction), and four in the tapering regions. Points  $B_1$  and  $B_3$  are some 3 mm from either end of the compressive taper while  $B_2$  and D are at the mid-points of the compressive and extensional tapers respectively. These positions are inherently somewhat imprecise due to the spatial divergence of the light beam as it propagates through the optically dense medium.

For a sample subject to a continuous, cumulative deformation, as here, the linear strain  $dl/l$  is a less appropriate measure than the Hencky (or natural) strain [19]

$$\varepsilon = \int_{l_0}^l \frac{dl}{l} = \ln(l/l_0), \quad (2)$$

where  $l_0$  is the original dimension of the sample. In this paper the term 'the strain' implies this Hencky strain. Similarly, the 'strain rate' implies the rate of change of the Hencky strain

$$\dot{\varepsilon} = \frac{1}{l} \frac{dl}{dt}. \quad (3)$$

Flow through a contraction, as in the present cell, is non-viscometric [19], and so cannot simply be related to simple shear flow. The strain is thus not easily expressible in terms of more fundamental quantities.

Values of  $\varepsilon$  and  $\dot{\varepsilon}$  appropriate to the various observation points are summarized in Table I. For point D  $l_0$  is taken as the width of the cell in region C, as the foam observed at D by DWS started out in that region. Both  $\varepsilon$  and  $\dot{\varepsilon}$  are necessarily somewhat imprecise because of the uncertainty in position of the observation points noted above.

The behaviour of foam is non-stationary [14, 15], so we studied the foam at a single age. At the highest flow rates used, we could not flow the foam continuously through the cell at a constant rate, due to the finite cell size (the barrier motion was limited to  $< 15$  cm). Thus the foam was allowed to age for some 150 min in the cell, then flow was initiated at the chosen rate. DWS observations were started at 165 min age, data being acquired over 15 min. Data from a single experiment at point  $B_3$  in which foam *was* continuously flowed through the cell at a fixed flow rate for several hours was in quite good accord with data for the same flow rate but

Table I. — Hencky strain and rate of change of Hencky strain (in terms of flow rate  $V$ ) at various observation points.

Position	$\varepsilon$	$\dot{\varepsilon}$
B <sub>1</sub>	-0.04	-0.15V
B <sub>2</sub>	-0.43	-0.33V
B <sub>3</sub>	-1.07	-1.20V
D	0.77	0.33V

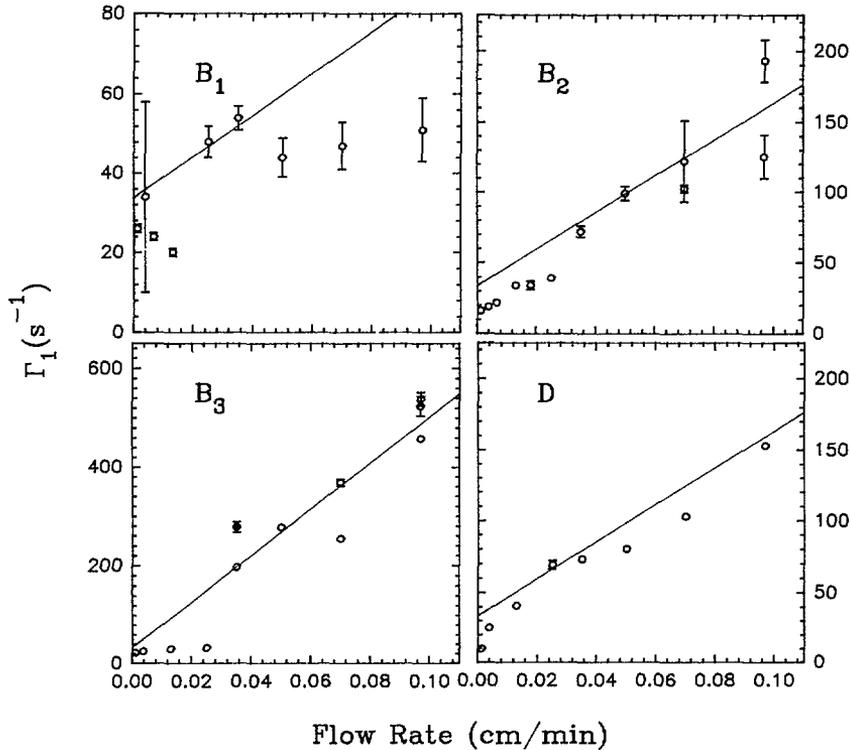


Fig. 2. — The variations of  $\Gamma_1$  with the flow rate (defined as the speed of the barriers in the cell, see text) at four points in the cell. The full point derives from an experiment in which foam was continuously flowed through the cell; the open points from experiments in which the foam was aged at rest before flow was induced. The line in each figure is that of the linear fit shown in Figure 3.

under the normal procedure (see Fig. 2). Each observation at a given flow rate and position derives from a separate sample of foam. Results from observations of static foam demonstrated the effects of aging, for comparison with the data for flowing foam.

### 3. Results and Discussion

We report here the decay constant  $\Gamma_1$  characterizing the initial decay of  $g_T^{(1)}(\tau)$ . Slower components, found in an earlier study [6], were observed, but will be neglected here. The decay constants of these slower components ( $\sim 4 \text{ s}^{-1}$ , apparently essentially independent of flow rate)

were significantly lower than any of the  $\Gamma_1$  data of the present study; the two components were thus easily separable, so that the fitted  $\Gamma_1$  values should not be affected by the slower decay. As we have noted elsewhere [6] the origin of these slow components is unclear, but we do not believe it is connected with the strain rate dependence of  $\Gamma_1$  which is of present concern.

DWS observations were made for flow rates from 0.0012 to 0.097 cm/min (Fig. 2). In some cases observations were repeated, the scatter of  $\Gamma_1$  being typically 10%. In these cases the uncertainties were taken as the standard error on the mean; otherwise the much smaller error on  $\Gamma_1$  from the data analysis was used.

We concentrate upon DWS observations in the tapered sections of the cell where the foam is subject to strain. In general, as we have noted elsewhere [7] in these regions  $\Gamma_1$  depends linearly upon the strain rate. However, there are significant departures from this linearity at low strain rate, upon which we focus in the present paper.

First we review the main features of the data, including the results of our previous work:

- The application of stress promotes foam rearrangement:  $\Gamma_1$  increases in the constricted foam [6].
- In regions of uniform flow (points A and C)  $\Gamma_1$  was essentially independent of the flow rate and was in accord with values for static samples [6].
- At the lowest strain rates investigated the  $\Gamma_1$  data from the tapered regions of the cell agreed with those for points A and C and for static samples.
- Between these very lowest values of  $|\dot{\epsilon}|$  and a (strain-dependent) critical value  $\Gamma_1$  systematically falls below the linear dependence just mentioned, being instead approximately independent of strain rate.
- Above the strain-dependent critical value of strain rate,  $|\dot{\epsilon}|$ ,  $\Gamma_1$  does not depend upon strain, but is simply linearly proportional to  $|\dot{\epsilon}|$  [7].

We now expand upon this brief summary to provide some context for understanding the present results.

Figure 3 shows all the  $\Gamma_1$  data (weighted means where appropriate) for observation points within the tapered regions of the cell as a function of the magnitude of the strain rate. Above the strain-dependent critical value of  $|\dot{\epsilon}|$  the data for points B<sub>2</sub>, B<sub>3</sub> and D do not depend upon strain, but rather are simply proportional to  $|\dot{\epsilon}|$  [7]. The straight line plotted in the figure represents a fit to the data for the four highest flow rates at points B<sub>2</sub> and B<sub>3</sub>, involving compressive (negative) strain. The data from point D, corresponding to expansive (positive) strain, accord excellently with this behaviour. The data for point B<sub>1</sub> never display this linearity.

In regions of uniform flow (or constant strain), and also at the lowest strain rates investigated in the tapered regions of the cell,  $\Gamma_1$  values were consistent with those found for static foam [6]. We assume that under these circumstances elementary rearrangements occur as coarsening (i.e., aging) leads to relaxation of local internal stress. At points A and C  $\Gamma_1$  was independent of the flow rate, the averages ( $16.6 \pm 2.3 \text{ s}^{-1}$  at A and  $13.1 \pm 1.7 \text{ s}^{-1}$  at C) being indistinguishable despite the non-zero (Hencky) strain at C. In these regions the observed  $\Gamma_1$  values are in accord with those for static samples [6], reflecting the plug nature of foam flow in a smooth-walled vessel [3]: apart from stick-slip of a boundary layer of bubbles, the foam flows as a solid object. Similarly, at the lowest strain rates investigated ( $\leq 0.0022 \text{ min}^{-1}$ ) the  $\Gamma_1$  data for all points in the tapered regions (weighted mean =  $21.8 \pm 2.0 \text{ s}^{-1}$ ) are in reasonable accord with those for points A and C, or for static foam. The intercept of the linear fit mentioned above,  $33 \pm 17 \text{ s}^{-1}$ , while somewhat higher than the value determined at points A and C, is also not inconsistent

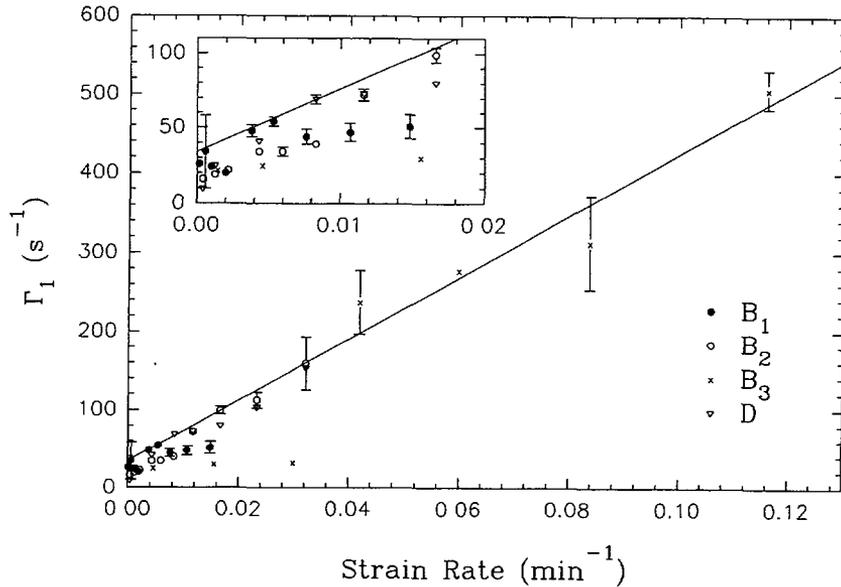


Fig. 3. — The dependence of  $\Gamma_1$  for points B<sub>1</sub> to B<sub>3</sub> and D upon the absolute value of the strain rate. The line is a linear fit to certain of the data; see text for details. The data at low  $|\dot{\epsilon}|$  are enlarged in the insert.

with the values at these points within the large uncertainty. At low strain rates the differences between simple plug flow behaviour and that expected from strain-enhancement (i.e., the linear dependence on  $|\dot{\epsilon}|$ ) are not large. It would thus be difficult to distinguish between solid-like behaviour (i.e., aging) and plastic flow here, even when the foam is subject to significant strain at certain points in the cell.

We now turn to the main point of present concern: the deviations of  $\Gamma_1$  from the linear dependence. Between the very lowest values of  $|\dot{\epsilon}|$  (where  $\Gamma_1$  agrees with the value for constant strain or for static foam) and the strain-dependent critical value (above which  $\Gamma_1 \propto |\dot{\epsilon}|$ ) rather different behaviour is apparent. This is particularly evident in the enlarged insert to Figure 3. In this intermediate range of strain rate the observed  $\Gamma_1$  for points B<sub>1</sub>, B<sub>2</sub> and B<sub>3</sub> systematically fall below the linear dependence found at larger  $|\dot{\epsilon}|$ , and instead are approximately independent of strain rate. Indeed, for point B<sub>1</sub> the data do not cross over to the linear dependence for any strain rate studied. For point D any change in behaviour at a critical  $|\dot{\epsilon}|$  is less than that for B<sub>2</sub> and B<sub>3</sub>, although there are some indications of a flattening of the variation of  $\Gamma_1$  with flow rate (and hence with  $|\dot{\epsilon}|$ ) about 0.04 cm/min (Fig. 2).

It appears that in the present experiments the simple linear dependence of  $\Gamma_1$  on the absolute strain rate [7] requires two conditions to be satisfied: a threshold strain must be exceeded *and*  $|\dot{\epsilon}|$  must be above a critical value. The threshold strain will just correspond to the yield stress for foam of gas volume fraction  $\phi = 0.92$  [3, 10, 22]. (To avoid possible confusion we will refer below to 'yield stress' rather than 'threshold strain'.) However, the requirement that the strain rate must exceed a critical value does not appear to emerge from current computer simulations. We may think of point B<sub>1</sub> as either lying below the yield stress or as never reaching the critical strain rate; given the limited experimentally accessible range of  $\dot{\epsilon}$  these alternatives cannot be separated.

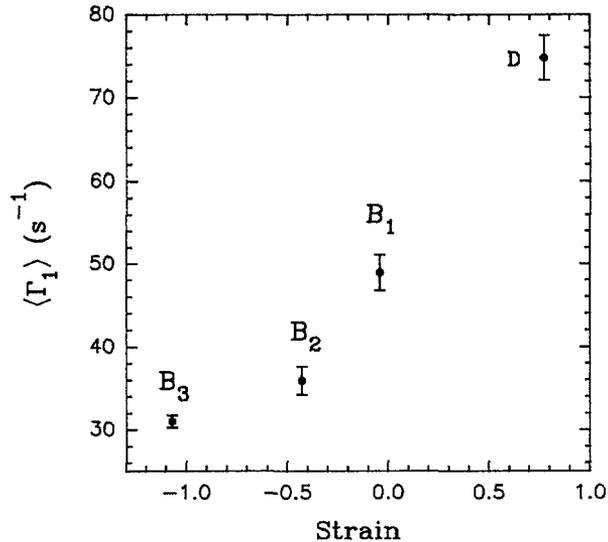


Fig. 4. — The strain dependence of  $\langle \Gamma_1 \rangle$  below the critical value of  $|\dot{\epsilon}|$ .

When the strain rate lies below the critical value at a particular strain  $\Gamma_1$  is relatively low and depends on  $\epsilon$ , not on  $|\dot{\epsilon}|$  (Fig. 3). Provided the foam is above the yield stress,  $\Gamma_1$  increases discontinuously as the critical strain rate is exceeded, thereafter following the linear dependence. As the magnitude of the negative strain increases, the value of  $\Gamma_1$  at low  $|\dot{\epsilon}|$  falls systematically (progressing from point B<sub>1</sub> to point B<sub>3</sub> in Figure 3). This is summarized in Figure 4, showing the average  $\Gamma_1$  from the region below the critical strain rate, as a function of the strain. The datum at positive  $\epsilon$ , from point D, is taken from the region of apparent constancy for flow rates about  $0.04 \text{ cm/min}^{-1}$  (Fig. 2); it seems to fit in well with the variation apparent for negative  $\epsilon$ .

The critical strain rate apparently depends on the absolute strain at a particular observation point (Fig. 3). Given the inherent uncertainties in both  $\epsilon$  and  $\dot{\epsilon}$  due to the spreading out of the light beam in traversing the sample we do not pursue the functional dependence here.

It is apparent that there is a real change in the behaviour of the foam at the critical  $|\dot{\epsilon}|$ ; below this critical strain rate  $\Gamma_1$  essentially depends only on the *strain*, whereas above this  $|\dot{\epsilon}|$  it increases linearly with strain *rate*. These effects are real: Figure 5 shows a correlation function observed at point B<sub>3</sub> for  $|\dot{\epsilon}|$  just below the critical value, together with the variation which would be expected if the linear variation extended to this strain rate. Clearly the observed correlation function does decay significantly slower than expected (observed  $\Gamma_1 \sim 30 \text{ s}^{-1}$ , expected value  $\sim 150 \text{ s}^{-1}$ ). We note that the difference could not arise from a slow component having decay constant  $\sim 4 \text{ s}^{-1}$ , justifying our belief that the present effects are not due to the slow components mentioned above.

Before discussing the origin of these effects at low strain rates, we review the understanding of the linear dependence of  $\Gamma_1$  upon  $|\dot{\epsilon}|$ . The only length scale in the system, the typical bubble size, must set the scale of both  $l^*$ , the transport mean free path of light in the foam, and  $r$ , the radius of the foam volume involved in a typical rearrangement event. At a given foam age the bubble size should be constant; all samples were aged for the same time (150 min) and so should have evolved similarly. Variations in  $\Gamma_1$  thus imply corresponding variations in

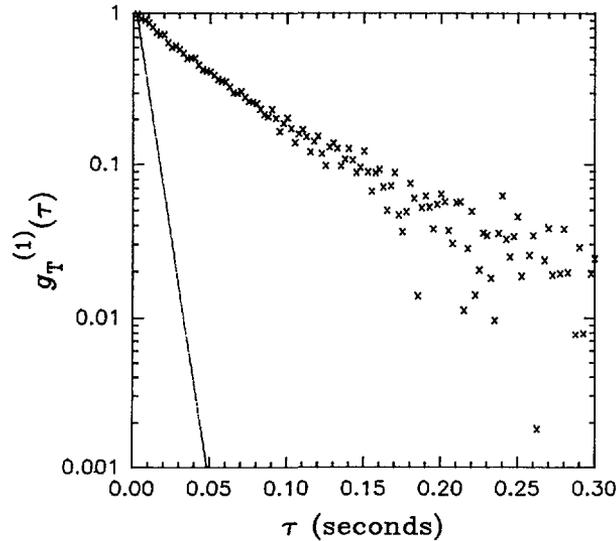


Fig. 5. — A DWS correlation function recorded at point B<sub>3</sub> with flow rate 0.025 cm/min. The line indicates the exponential correlation function expected if  $\Gamma_1$  were as predicted by the line in Figure 3.

$\tau_0$ , the average time between rearrangements at any specific location in the foam. The linear strain-rate dependence of  $\Gamma_1$  (Fig. 3) thus implies that  $\tau_0^{-1}$  is also linearly proportional to the strain rate [7].

The nature of the rearrangement events in flowing foam is not entirely clear. For static foam [14, 15] microscopic observation of the rearrangements in the surface of foam in contact with a glass wall suggested that foam coarsening leads to local build up of stress which is eventually relieved by a rearrangement of a foam volume some ten bubbles across. For static foam the rate at which this occurs was shown to be compatible with measured values of  $\Gamma_1$  [14]. For the case of flowing foam, simulations [9, 10, 21, 22] suggest that, above the yield stress, local stresses are relieved in a broadly similar manner; some work suggests that avalanches of rearrangement events spanning all length scales may occur [22].  $\Gamma_1$  carries information only on the probability of a photon being affected by a rearrangement event while traversing the sample [4, 5], and so our data cannot distinguish such details. However, it is evident that at a given strain there is a change in behaviour as the critical strain rate is exceeded.

The linear variation found above the critical strain rate accords well with the findings of recent computer simulations of the effects of stress upon two-dimensional foam: it seems that the picture just sketched is the appropriate one. However, there are no hints in the simulations of the strain dependence of  $\Gamma_1$  for  $|\dot{\epsilon}|$  below a critical value. It seems unlikely that this can be ascribed to elastic behaviour, as the simulations suggest that the foam responds elastically when the strain lies below a threshold value (corresponding to the yield stress). The conclusion that the present results are an unexpected feature of the plastic flow of foam is supported by the fact that the region of apparent constancy of  $\Gamma_1$  for point D (see Fig. 2) lies very close to the linear dependence (the line in the figure): for large positive  $\epsilon$  there is a smooth cross over from the low strain rate behaviour of  $\Gamma_1$  to the linear dependence upon  $|\dot{\epsilon}|$ .

In these experiments some mechanism, not apparent in the simulations, must be operative at the lower strain rates. In foam DWS samples spatially or temporally rare events: if  $\Gamma_1$

(and hence  $\tau_0^{-1}$ ) is lower than expected, it suggests that either  $R$ , the rate per unit volume of events de-phasing the light, or  $r$ , the typical size of such events, is smaller [14]. It does seem reasonable that at very low strain rates the processes to relieve the strain should be either rarer or on a smaller scale than at higher  $\dot{\epsilon}$ . The difference between the two possibilities is not very material at present: what matters is that there is a change at the critical  $\dot{\epsilon}$ . The plastic flow at high strain rates gives way to a quasi-static behaviour at low  $\dot{\epsilon}$ . What are the elementary events governing the DWS signal below the critical strain rate?

At this stage we can offer only qualitative arguments. Consider an elementary slab of foam moving through the cell. In region B the slab will be compressed laterally, as the flow is constricted. Bubbles will be forced closer together, and will wish to rearrange to relieve the stress. The only way for this to happen in this region is for bubbles to move back against the flow, the foam in front of our elementary slab being under even greater compressive stress. On the other hand, as the foam is being expanded in region D, the strain will be such that bubbles will be able to move forward from our elementary slab into more expanded layers ahead, being pushed by the more compressed foam behind. In region D the situation is not dissimilar to the growth of a flower such as a daisy, in which florets form a structure of more or less concentric rings; florets originate in the centre of the flower and find it easy to move out into the outer rings [20].

Such a process has not been observed in any computer simulations of foam rheology of which we are aware, but does seem to fit the present results. In region B the hypothesized stress relief events will be inhibited by the compressed nature of the foam ahead of the elementary slab and the pressure of the foam behind pushing forward, this inhibition increasing as the foam becomes more highly compressed, while in region D they will be comparatively easy, becoming steadily easier as the foam becomes more expanded. The strain dependence of  $\Gamma_1$  shown in Figure 4 thus emerges naturally from this scenario. In effect this argument links the present phenomena to the compressibility of the foam. At a given strain,  $\Gamma_1$  does not change with strain rate, reflecting constancy of  $\tau_0$ . The rate of de-phasing events per unit volume is thus also constant, assuming that the typical size of the events remains constant. Increasing  $|\dot{\epsilon}|$  (below the critical value) will increase the rate at which bubbles attempt to move back against the flow, but will also increase the inhibition of such movements just discussed. This interplay between the two influences may underlie the rough constancy of  $\Gamma_1$  below the critical strain rate. At strain rates above the critical value the build-up of stress will be more rapid and the more conventional rearrangements, occurring at a rate dependent on  $|\dot{\epsilon}|$  will predominate.

#### 4. Conclusions

In conclusion, while further work is needed to pursue some aspects of the present data, it is clear that the behaviour of foam in constrictive flow exhibits an unexpected strain-rate dependence. Below a strain-dependent critical strain rate the rate at which elementary bubble rearrangement events occur at any given point in stressed foam depends only on the strain, whereas above this critical value it is just proportional to the strain rate. This strain dependence of the rate of stress-relief events is not apparent in computer simulations to date.

Why do simulations not reveal the effects observed? Several possible reasons come to mind. The constrictive strain used in our experiments differs from the extensional [9, 10] or shear [21, 22] strains studied in simulations. The simulations typically involve a constant strain rate applied over time, to accumulate strain greater than the threshold value for plastic flow. This differs from our experiments where, at a given point in the taper of the flow cell, the strain is fixed and the strain *rate* is the experimental variable. Possibly more significantly, the simulations do not involve compression of the foam, whereas in the experimental flow the foam

is subject to a strain which is, in part, compressive. Further, the foams used in simulations have, with a few exceptions [12], been dry; the wetness of the experimental material may cause differences; in wet foams the basic topological processes familiar from studies of dry foams apparently do not occur, and different strain-relief mechanisms may appear. The systems studied in simulations have tended to be relatively small (few hundred bubbles) and two-dimensional, which again might tend to inhibit the appearance of certain processes. Finally, the imposition of periodic boundary conditions in the simulations places constraints on the system which are absent in our experiments. It is to be hoped that future work will elucidate which, if any, of these differences underlies the experimentally observed phenomena not so far seen in simulations.

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