corresponds with the need to invoke a spin polarization of the current in the normal metal of about 100%, to explain the results of refs 13 and 14 in terms of spin accumulation. In our opinion this must imply that the observed effects of refs 13 and 14 cannot be related to spin accumulation.

We have thus demonstrated spin injection and accumulation in a mesoscopic spin valve. We find a surprisingly long spin-flip length in Cu of around 1 μm at 4.2 K and about 350 nm at room temperature. For the smallest Py electrode spacing, the magnitude of the spin signal and the current polarization P in the Cu wire are limited by the unfavourable ratio of the spin-independent resistance of the Cu strips (L/σ_N) and the spin-dependent resistance of the Py ferromagnet (λ_F/σ_F). In principle, larger signals can therefore be obtained by a proper choice of materials and geometries.

Finally we note that our system permits the study of spin transport phenomena, such as controlled spin precession in solid state devices and the control of spin-polarized currents at room temperature by additional ferromagnetic contacts^{3,26}. □

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Shear-induced molecular precession in a hexatic Langmuir monolayer

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Liquid crystalline behaviour is generally limited to a select group of specially designed bulk substances. By contrast, it is a common feature of simple molecular monolayers and other quasi-two-dimensional systems¹, which often possess a type of in-plane ordering that results from unbinding of dislocations—a ‘hexatic’ liquid crystalline phase. The flow of monolayers is closely related to molecular transport in biological membranes, affects foam and emulsion stability and is relevant to microfluidics research. For liquid crystalline phases, it is important to understand the coupling of the molecular orientation to the flow. Orientationally ordered (nematic) phases in bulk liquid crystals exhibit ‘shear aligning’ or ‘tumbling’ behaviour under shear, and are described quantitatively by Leslie–Ericksen theory². For hexatic monolayers, the effects of flow have been inferred from textures of Langmuir–Blodgett films^{3–5} and directly observed at the macroscopic level^{6–10}. However, there is no accepted model of hexatic flow at the molecular level. Here we report observations of a hexatic Langmuir monolayer that reveal continuous, shear-induced molecular precession, interrupted by occasional jump discontinuities. Although superficially similar to tumbling in a bulk nematic phase, the kinematic details are quite different and provide a possible mechanism for domain coarsening and eventual molecular alignment in monolayers. We explain the precession and jumps within a quantitative framework that involves coupling of molecular orientation to the local molecular hexatic ‘lattice’, which is continuously deformed by shear.

The interfacial thermodynamics and rheology of molecular films like Langmuir monolayers are responsible for the stability of multiphase materials such as foams and emulsions¹¹. They are important model systems for studies of dynamics and molecular interactions in bio-membranes and, as quasi-two-dimensional systems, display a variety of unusual liquid crystalline phenomena. The application of synchrotron X-ray scattering and specialized imaging techniques such as fluorescence and Brewster angle microscopy (BAM) have

Figure 1 Hexatic and nematic phase representations. **a**, A cartoon showing the molecular geometry of rod-shaped molecules in a tilted hexatic phase. The molecule is tilted away from the surface normal by the angle θ , and the azimuthal direction of tilt is given by the angle ϕ . **b**, Schematic picture of a tilted hexatic phase viewed from above. The projection of the molecules on the surface plane are oval; the dark circles represent the headgroups at the interface. In this case, the molecular tilt direction is locked into the nearest neighbour direction of the local distorted-hexagonal unit cell. We note that the orientation of the unit cell remains uniform despite the presence of a lattice dislocation that destroys the positional registry of the lattice. **c**, Schematic picture of a nematic phase. Molecules are aligned in a common direction on average, but their positions are random.

greatly improved our understanding of monolayer structure and phase behaviour in the last decade¹². Hexatic phases were found to be extremely common in monolayers and, in fact, characteristic of these systems. However, a variety of recently discovered dynamic and rheological phenomena, such as the coupling of molecular orientation to flow^{6–10,13,14}, unusual relaxation dynamics and shear moduli^{15–17}, and non-newtonian velocity profiles in channel flow^{18–20}, remain largely unexplained. Another mysterious effect of flow is that it may have either an annealing or a fragmenting effect on the monolayer domain structure (L'_2 and Ov phases respectively^{6,7}).

Many hexatic monolayer phases are characterized by a finite molecular tilt at a polar angle, θ , with respect to the surface normal (see Fig. 1a). A typical monolayer sample consists of a mosaic of macroscopic domains separated by sharp boundaries; molecules within a given domain share a common azimuthal tilt direction, φ (Fig. 1a). In these phases, the tilt direction is directly linked to the orientation of the local pseudo-hexagonal unit cell (Fig. 1b). The orientation of the unit cell extends over macroscopic distances, although true crystalline order is destroyed by the presence of local point defects (lattice dislocations). The molecules in a nematic phase (Fig. 1c), on the other hand, are aligned on average in a particular direction; however, their positions are completely disordered. The coupling of molecular orientation to flow is well understood for bulk nematic phases², and is important in the processing of many polymeric and/or fibre-based materials. There is evidence that the description of nematic flow can be extended to two dimensions. Workers^{21,22} who studied the transient behaviour of a nematic monolayer (of a hairy-rod polymer) under extensional flow found good agreement with a two-dimensional rigid rod model. There is no general understanding or accepted picture of the coupling of structure to flow in hexatic phases.

Because the intensity (grey level) within a BAM image is directly related to φ , BAM provides a convenient method of studying changes in the molecular orientation. Previous observations in fatty acid monolayers suggested that flow induced a reorientation of the molecules to a particular angle^{6–8} with respect to the flow. The mechanism for this “alignment” was the sudden appearance of new domains with the appropriate orientation. The domains grew by movement of a boundary; the propagating fronts were described as shear bands^{6,7}. We report a qualitatively different phenomenon: a continuous change in the reflectivity of monolayer domains corresponding to a cooperative, continuous rotation of the molecular orientation.

Figure 2 shows a time sequence of BAM images corresponding to a simple shear flow experiment⁸ on a docosanoic acid

($\text{CH}_3(\text{CH}_2)_{20}\text{COOH}$) monolayer in the L'_2 phase¹² ($T = 17^\circ\text{C}$). Before the onset of flow (Fig. 2a), the central fish-shaped domain is darker than the surrounding region. As shear is initiated (Fig. 2a, b) the shape of the domain is gradually skewed and the contrast is reduced between the domain and the surroundings. At some point the contrast is almost completely lost (Fig. 2d). As shear continues, the original domain reappears (with a distorted shape), but the contrast has been inverted. Qualitatively, this sequence corresponds to a simultaneous anticlockwise rotation of the molecules within the central domain from left to right and of the molecules in the surrounding region from right to left. This particular example of contrast inversion is clear evidence of continuous molecular precession. However, neighbouring domains do not generally rotate simultaneously, or in phase with each other. The independence of domain dynamics suggests that the precession is not related to domain–domain interactions. Our observation, in recent experiments, that domains can ‘slip’ along each other during shear is also consistent with a picture where the molecular details of neighbouring domains are de-coupled.

We have measured the evolution of the grey level during 35 precession events. In all cases, after what can be long periods of time in an initial state where the tilt azimuth is approximately parallel to flow, the grey level of a domain changes relatively quickly from dark to light (corresponding to a change in molecular orientation from the left to the right of the incidence plane), or vice versa. This corresponds to a nearly 180° rotation of the molecules, after which the grey level remains essentially constant again. Representative data presented in Fig. 3 show that a higher shear rate ($\dot{\gamma}$) results in faster molecular precession (Fig. 3a). In fact, the rate of change of the reflectivity is proportional to the shear rate (see Fig. 3b) and the data can be collapsed when plotted as a function of the strain ($\gamma = \dot{\gamma}t$). This indicates a flow-induced reorientation of the alkane tails. The scaling with shear rate also suggests that our observations are made under steady state conditions, where elastic effects can be ignored. The qualitative behaviour, that is, fast change in reflectivity followed by saturation (more clearly observable in Fig. 4) is reminiscent of a Jeffery orbit²³, which describes the rotation of an elongated object under shear or the tumbling of nematic molecules. The Jeffery orbit is periodic and predicts that the domain will eventually return back to its original intensity. A quantitative comparison, however, reveals that the observed dynamics cannot be reconciled with a Jeffery orbit. In particular, a Jeffery orbit that is consistent with the steepest region of the curves in Fig. 3 predicts that the intensity will pass a maximum and start to decrease well within the long period of time during which the observed intensity remains constant. Our observation of asymptotic regimes before and after rapid change instead

Figure 2 Continuous change in the reflectivity of domains in a docosanoic acid ($\text{CH}_3(\text{CH}_2)_{20}\text{COOH}$) monolayer in the L'_2 phase. ($\Gamma = 23 \text{ mN m}^{-1}$). Shear is counter-clockwise ($\dot{\gamma} = -0.2 \text{ s}^{-1}$) and the flow field is horizontal. Images are obtained with a BAM with the laser incident from the top (perpendicular to the flow) and the analyser set at 60° for optimal contrast. Scale bar in **a** is $100 \mu\text{m}$ long. The grey level of the images is linearly related to the local reflectivity. Qualitatively, the molecular azimuth in the dark domains is oriented towards the left of the incidence plane and the light domains towards the right. Before this sequence, the monolayer had been prepared through a series of shear reversal cycles (in which the flow is inverted after half a cycle), resulting in the

reduction of the initially large number of reflectivities in the mosaic of domains to only two different reflectivities. The dark domain in the centre of **a** exhibits a continuous increase of its reflectivity, while the surrounding domain darkens as strain increases corresponding to continuous anticlockwise precession of the molecular orientation in both domains. Elapsed times after frame **a** are as follows: **b**, $t = 1.7 \text{ s}$; **c**, $t = 2.9 \text{ s}$; **d**, $t = 4.3 \text{ s}$; **e**, $t = 5.4 \text{ s}$; and **f**, $t = 6.8 \text{ s}$. The contrast drops to zero between frames **c** and **d** as the reflectivities of both domains become equal. Optical artefacts such as interference fringes and illumination gradients are present and unavoidable.

of extrema followed by periodic behaviour indicates a clear incompatibility with the nematic tumbling model.

Our experiments also reveal the presence of jump discontinuities in the otherwise continuous change in the reflectivity of the monolayer (Fig. 4). They appear as a diffuse front that propagates in arbitrary directions, changing the reflectivity of the affected domain (this is clearly different for the sharp and well-oriented alignment fronts observed in other monolayer phases). In many instances, the magnitude of these jumps is close to 30° , as discussed below.

Both the presence of discontinuous orientational jumps and the quantitative details of the precession dynamics are inconsistent with expected behaviour for a tumbling nematic phase. Although we can imagine the projection of a tilted molecule onto the surface plane as an analogue of the nematic director, the structural details of hexatic monolayer phases are quite different from those of a nematic. In particular, there is a strong coupling between the tilt azimuth and the orientation of the local pseudo-hexagonal unit cell, the hexatic bond-orientational order parameter^{12,24,25}. Both L'_2 and O_v phases have molecules tilted towards their next nearest neighbour (NNN) while the L_2 phase is characterized by a tilt towards a nearest neighbour (NN) molecule. An attempt to describe the dynamics of these phases using a nematic model completely neglects the existence of hexatic order and the coupling of the molecular tilt to the local lattice.

Although shear flow continuously distorts the shape of the underlying lattice, we expect that the tilt azimuth of a given molecule will prefer to remain in a position that approximates the NNN environment. This is consistent with our observation that cessation of the flow at any time during shear gives a configuration whose domain reflectivities do not appear to relax with time. As a first-order model, we propose that the tilt azimuth, φ , of a given molecule attempts to follow the displacement of its NNN as the lattice is

Figure 3 Evolution of the 8-bit digitized reflectivity for three different domains under three different shear flows. The monolayers are in the L'_2 phase. ($\Pi = 20 \text{ mN m}^{-1}$). **a**, The mean (data point) and the standard deviation (error bar) of the instantaneous distribution of grey levels of the domain under observation are shown combined. **b**, The same data are shown after the time axis is transformed into the strain axis. The data sets are superimposed by adding arbitrary offsets to the strain axis.

deformed by shear (Fig. 5). This leads to the relationship $\cotan[\varphi(t)] = \dot{\gamma}(t - t_0)$, with $t_0 = -x_0/(\dot{\gamma}y_0)$. This expression is qualitatively consistent with our observation of a steep change in the reflectivity of the domains preceded and followed by asymptotic regimes (Figs 2 and 3). This continuous model, however, is topologically inconsistent with the NNN lattice-bond symmetry condition. Indeed, as the lattice is distorted by shear, the relative position of different sites will change and sites that originally were NNN actually become NN (see Fig. 5); not a stable tilt direction in the L'_2 phase. The observed jump discontinuities in the evolution of $\varphi(t)$ resolve this inconsistency. Composing the expression for $\varphi(t)$ with the calculated reflectivity as a function of φ , we obtain an expression that is fitted to the data in Figs 3 and 4. In this way, we

Figure 4 Jump discontinuities are revealed in the reflectivity of two domains from different experiments shearing a monolayer in the L'_2 phase. **a**, $\Pi = 22 \text{ mN m}^{-1}$, $\dot{\gamma} = 0.2 \text{ s}^{-1}$ (clockwise); **b**, $\Pi = 20 \text{ mN m}^{-1}$, $\dot{\gamma} = -0.05 \text{ s}^{-1}$ (anticlockwise). The solid lines are fits to the data using the model proposed in the text and that characterize the jumps as follows: **a**, $\varphi = 311^\circ \pm 7^\circ$ to $\varphi = 278^\circ \pm 7^\circ$ ($\Delta\varphi = -33^\circ \pm 10^\circ$); **b**, $\varphi = 262^\circ \pm 6^\circ$ to $\varphi = 294^\circ \pm 6^\circ$ ($\Delta\varphi = 32^\circ \pm 9^\circ$). Our 35 observed precession events have clearly revealed 23 instances of jump discontinuities. We have obtained a quantitative estimation for the magnitude of the jump in 17 of those cases, which resulted in 11 jumps of magnitude close to 30° .

estimate the magnitude of the experimentally observed jump discontinuities, many of which are close to 30° .

Figure 5 provides a geometrical interpretation for the observed magnitude of the largest jumps, which we are most likely to observe. The jump depicted in Fig. 5 has a magnitude of 26° within an angular range where the variation in BAM intensity is large, consistent with the majority of our observations. This picture also predicts that a number of smaller jump discontinuities ($<15^\circ$) will continue to occur as large strains are applied in order to reconcile the NNN lattice-bond orientation and the lattice distortion. These jumps would take place in a region of reduced reflectivity and would be smaller in magnitude, making them more difficult to observe.

We believe that the macroscopic appearance of monolayers following flow is directly related to the coupling between flow and molecular orientation. The combination of continuous precession and jump discontinuities of molecular orientation in the L_2' phase allows gradual alignment of molecular orientation in adjacent domains, resulting, ultimately, in a coarsened domain structure with two predominant molecular orientations. However, the

nucleation and propagation of new domains upon initiation (or reversal) of flow in the Ov phase breaks up previously coherent domains, leading to increasing fragmentation.

The details of the molecular precession reported here combined with our previous observations regarding shear alignment of the molecular lattice in the Ov phase⁸ suggest a conceptual framework for the flow of hexatic phases that is fundamentally different from that for nematic phases. Although qualitatively similar phenomena are observed, that is, shear-alignment and continuous orientation rotation, they are distinctly different when analysed quantitatively. Flow acts directly on the molecular orientation (director) in a nematic phase whereas the local unit cell orientation dominates the behaviour in a hexatic monolayer; the molecular azimuth is essentially locked into the lattice. This concept can be used to guide our thinking about the effects of flow in quasi-two-dimensional systems (monolayers, biological membranes, and so on), in which hexatic phases are so prevalent. □

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Figure 5 Geometrical model for the onset and magnitude of the jump discontinuities in the evolution of $\varphi(t)$. This picture assumes that the degenerate lattice lines align with the flow. This is the same lattice alignment that was observed as a result of shear alignment in the Ov phase⁸ and is consistent with the observations reported here. The flow field (horizontal clockwise shear) relative to the reference site (1) is illustrated by the arrows in **a**. **a**, A region that initially satisfies the next nearest neighbour (NNN) lattice-bond orientation is considered. The azimuth in the reference site (1) is towards a NNN (2), whose initial coordinates with respect to (1) are (x_0, y_0) . These coordinates change to $(x_0 + \gamma t y_0, y_0)$ when a strain γt has been applied. **b**, After a strain of $\gamma = 0.6$ the continuous reorientation results in a violation of the NNN condition, as (1) and (2) now become NN. **c**, A jump in $\varphi(t)$ can result in a more favourable configuration, closer to the NNN, with (1) tilted towards (3). In this example, the jump takes place between $\varphi = 310^\circ$ and $\varphi = 284^\circ$ ($\Delta\varphi = -26^\circ$), consistent with the data in Fig. 4a.

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