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Dynamic Simulations of Potentially Auxetic Liquid-Crystalline Polymers incorporating Swivelling Mesogens

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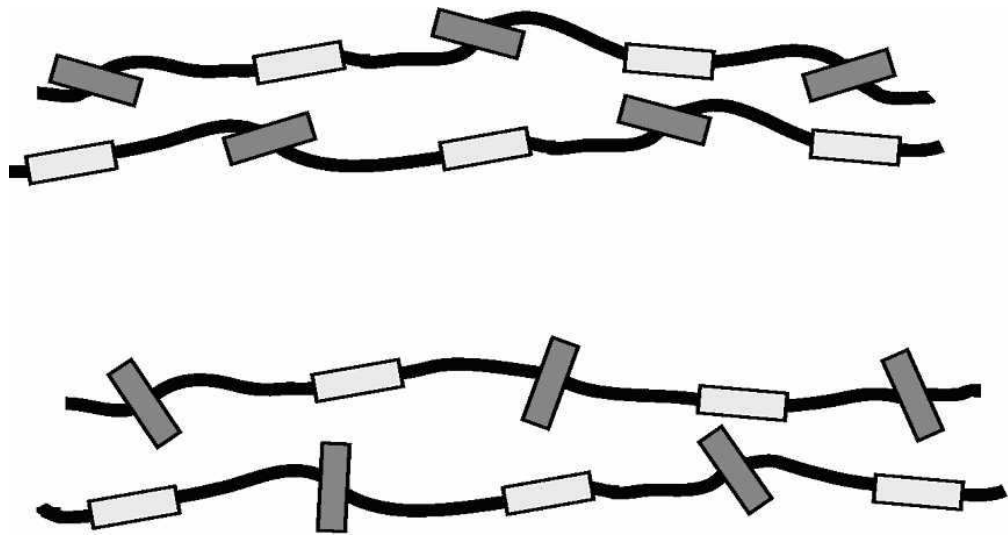


Figure 2 Liquid crystalline polymer structure containing swivelling mesogens before and after stretching
228x120mm (96 x 96 DPI)

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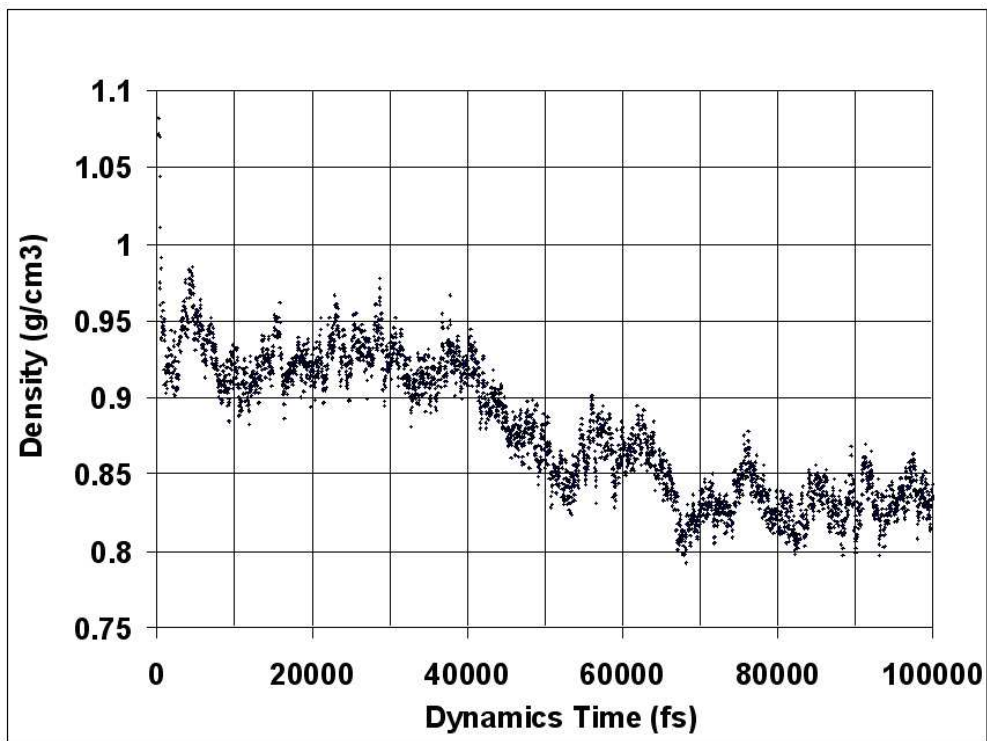


Figure 6 Density versus time for a dynamic simulation run of polymer S7 under a stress of 0.15 Gpa.
228x171mm (96 x 96 DPI)

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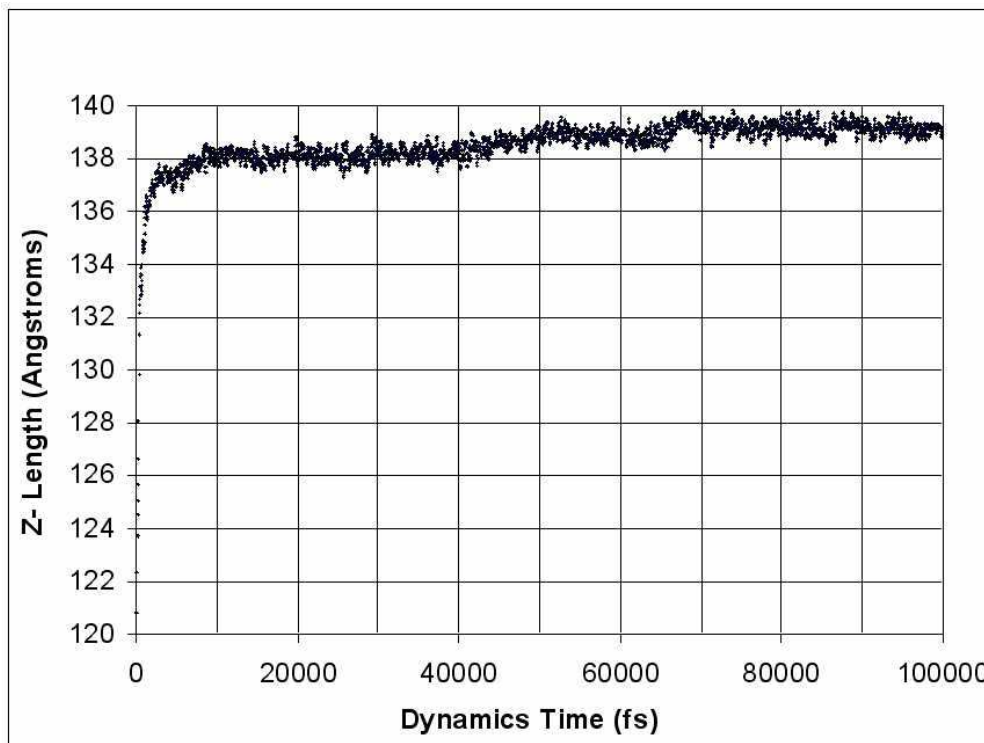


Figure 7 Z-axis length versus time for a dynamic simulation run of polymer S7 under a stress of 0.15 Gpa.
228x171mm (96 x 96 DPI)

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3 Dynamic Simulations of Potentially Auxetic Liquid-Crystalline Polymers incorporating
4 Swivelling Mesogens
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10 P. Aldred and S. C. Moratti*

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17 Cambridge, Cambridge CB2 1EW, UK.
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19

20 **Abstract**
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24 Dynamic simulations under stress were carried out on a series of main-chain polymers
25 containing two types of mesogen. One mesogen is orientated in-line with the chain (end-
26 to-end), the second (swivelling) has the main chain passing at right angles through it.
27 Under strain the swivelling mesogens are expected to rotate, pushing apart neighbouring
28 chains. Polymers incorporating 50% of both mesogens showed a reduced Poisson's ratio.
29 In particular, **swivelling** mesogens of seven rings length produced a negative Poisson's
30 ratio, and thus should be auxetic. In contrast, polymers containing only **end-to-end**
31 mesogens had relatively high Poisson's ratios of 0.53-0.59.
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46 **Key words**
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50 Dynamics simulation, liquid crystalline, negative poisson ratio, auxetic, polymer.
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Main Text

Most known auxetic materials (i.e those with a negative Poisson's ratio) owe their unusual behaviour to relatively large-scale deformations within their structure (for reviews of auxetic materials see ref. [1-2]. These materials include re-entrant foams, expanded fibres, and unusual honeycomb structures. Several crystalline materials are known that are believed to be auxetic, including cristobalite [3]. However, they are often only auxetic along one or more crystallographic axis. It would be potentially very useful to discover a polymer that has intrinsic auxetic properties. Auxetic polymer materials have been synthesised by careful particle sintering [4, 5], however the auxetic behaviour arises from the fusion and subsequent deformation and motion of macroscopic particles rather than the motion of individual polymer chains.

Several molecular polymeric structures have been proposed as being potentially auxetic. Evans and coworkers [6] proposed a very rigid re-entrant honeycomb based on phenylacetylene units (figure 1). While such a structure was shown to be auxetic by force-field type calculations, it is not likely to be easily synthesised or processed. As well, it is auxetic only in one plane, and a bulk material consisting of randomly oriented crystallites of structure 1 has is not necessarily going to be auxetic. Another structure put forward by Wei was based on hydrogen-bonded polymer network [7] . This hypothetical polymer had the advantage of being processible. While molecular mechanics-based

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3 calculations indicated auxetic behaviour, the H-bonded structure actually calculated was
4 only one of many possible. As well, the material was only auxetic in one plane as before.
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10 A useful, intrinsically auxetic, polymer would have to have a negative Poisson's ratio in
11 more than just one plane for it to exhibit bulk auxeticity. In its simplest representation it
12 would be an amorphous material. A highly aligned single chain polymer structure might
13 also be possible, however the practical difficulty of aligning non-liquid-crystalline or
14 non-crystalline polymers should not be underestimated. One problem intrinsic to
15 amorphous non-crosslinked polymers is that much of the effective molecular deformation
16 behaviour under strain may involve Van der Waals and non-bonded contacts as much as
17 covalent bonds. Thus, designing a linear covalent system to transfer strain into a
18 perpendicular direction might be defeated by non-covalent slippage and some unwanted
19 deformations of the main chain.
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36 Griffin and co-workers suggested a novel route to main chain auxetic polymers. This
37 involved taking advantage of the natural alignment of rigid mesogens, and using of
38 mixture of end-to-end and laterally-attached (swivelling) linkers [8, 9]. One big
39 advantage of this system is that in principle, (as it is liquid crystalline), it should be much
40 easier to align the chains, leading to more covalent than noncovalent bond force transfer.
41 It was predicted that under no strain the polymer would exist in a conformation with all
42 the mesogens aligned, however that under an applied stress the laterally attached
43 mesogens would swivel, forcing the chains apart. Experimental results have not yet
44 conclusively shown auxetic behaviour, but there have been promising indications using
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3 x-ray scattering, where interchain spacing was shown to increase on either macroscopic
4 alignment [8] or under strain [9].
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10 The systems used by **Griffin** et al was not ideal in that the degree of swivelling may be
11 limited by the laterally attached linking groups. They used two different linking
12 strategies. The first was a simple phenyl ether, the second a biphenyl ether. They
13 suggested that the first was limited to an swivelling angle of 60° (figure 3a), and the
14 second to a slighter larger angle of 75° (figure 3b) [9]. However, these figure would need
15 to be confirmed by more rigorous modelling studies, as much would depend on the
16 starting conformation, which may not be exactly at exactly 0°.
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29 We decided to take a preliminary look into the modelling of these systems. This was
30 thought very useful as the synthesis of potential candidate systems is not always
31 straightforward and the modelling may allow one to concentrate on promising leads. We
32 decided to use another linking strategy based on a cyclobutadiene system that should
33 allow a swivelling angle of almost 90°, (figure 3C) increasing the chances of finding an
34 auxetic structure. It should be noted that the particular ring system chosen is probably not
35 very chemically stable or synthetically friendly as such, but similar alternatives can
36 probably be found in practice.
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50 The proportion of swivelling and end-to-end mesogens was initially set at 50:50
51 (although again in practice slightly more end-to-end mesogens might have to be
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3 employed to ensure the right alignment). This again was to increase the chance of finding
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5 auxetic behaviour.
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10 There are several possible ways of computing the Poisson's ratio from an atomistic level.
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12 In the Cerius[®] suite of programmes there are two in-built methods. The static deformation
13 method [10] involves performing 12 deformations - three pairs in uniaxial
14 tension/compression and three pairs involving pure shear -- followed by a reminimization
15 to restore a state of detailed mechanical equilibrium. Each of these deformations
16 corresponds to setting one of the components of the strain vector to some small value
17 (e.g., $\epsilon = 0.001$), while keeping all other components fixed at zero. The elastic stiffness
18 coefficients can then be obtained by estimating the second derivatives of the deformation
19 energy with respect to strain using a finite difference formula. The other method for
20 determining the Poisson's ratio is by the fluctuation method [11]. Here molecular
21 dynamics simulations (NPT ensemble) are run and from the resulting saved frames the
22 correlation between stress and strain is established over the time period of the simulation.
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24 It has been showed that at least 10^5 samples need to be collected before the values for the
25 elastic constants have converged.
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48 These methods are appropriate to the study of highly crystalline systems under low strain,
49 in which any small stress-strains are efficiently transmitted to the perpendicular direction.
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51 The problem with the main chain polymers here is that small strains might be
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53 accommodated by small changes in the main chain conformation or by other, non-
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3 covalent, effects. It is not until higher strains that any swivelling might be expected to be
4 felt (Figure 4). Because of this, it was felt more appropriate to use molecular dynamic
5 simulations under an applied stress [12, 13]. In this way, much higher strains, of up to 10-
6 30%, could be reasonably investigated.
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15 Three polymers structures were looked at, varying the swivelling mesogen length. Of
16 course, there are a large range of parameters that can be investigated, such as chain length
17 and composition, swivelling mesogen concentration, co-monomers etc. The mesogen
18 length seemed the most important factor, and the one that influences all aspects of the
19 properties of the liquid-crystalline polymer.
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32 **Experimental**

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36 All of the modelling studies carried out in this project used Accelrys® 'Material Studio'
37 Modelling version 3.0 [14] running on an Intel Pentium - 4 workstation (with 2 x 3.20
38 GHz parallel processors and 512 MB RAM for internal memory). The molecular
39 dynamics runs were run unless otherwise at 1fs/step, using an Anderson [15] thermostat,
40 and a Parinello-Rahman [16] Barostat.
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51 1). The polymer cells were first constructed by making an alternating copolymer with a
52 orthogonal periodic cell of 5 chains, each consisting of 5 units of the structures shown in
53 figure 5. These were packed at low density using the Visualiser module. A molecular
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3 dynamic run under a moderate barostatic pressure (0.05 Gpa, 298 K) forced the chains
4 together to a realistic density.
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8 2). This was then equilibrated at NPT (no applied pressure) conditions using molecular
9 dynamics at 500 K for at least 5000 steps to get rid of any unfavourable high energy
10 conformations. The cell was then minimised under the Compass forcefield [17, 18], and
11 then equilibrated to constant volume under molecular dynamics (NPT, 298 K) for 50-100
12 K steps (1 fs each). **The Compass forcefield was specifically developed with**
13 **condensed phases and polymers in mind and was seen as a good starting point.**
14 **However, other forcefields may well give slightly different results, and this will be**
15 **looked at in future studies.** The densities achieved (1.0-1.15) and the order parameters
16 of the mesogens (0.6-0.74) appeared realistic. It was found that if in practice that unit cell
17 angles at this stage departed too much from 90°, then the next stage (dynamics run under
18 applied stress) often failed due to very large scale shear deformations. To avoid this, if
19 the cell angles were more than about three degrees from ninety, then the structure was
20 reinserted into a orthogonal unit cell of the same volume and step 2 repeated.
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41 3). Another molecular dynamics run was performed at 298 K, this time under stress in the
42 Z (chain) direction until a constant strain was reached. The precise stress applied
43 depending on the particular polymer studied, but to get a strain of 0.05-0.2 a stress of 0.1-
44 0.2 Gpa was needed. This equilibration typically took 50-100K steps. With this
45 optimisation, it is important to check the final structures as occasionally a high energy
46 structure was obtained. **This usually happened when a chain or mesogen got trapped**
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3 **under stress, and could be seen by the presence of a bent biphenylene structure.** If
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5 this occurred the process was repeated with a slightly different starting conformation.
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8 4). The resulting unit cell volume was then compared to the starting unit cell. As the
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10 material should be in principle isotropic around the z-axis, then there is no need to try and
11
12 calculate individual zx and zy Poisson ratios (in practice there are small differences in the
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14 present models, but this is due to the relatively small unit cell size). Instead the Poisson's
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16 ratio (ν) for the z-direction was calculated by the overall change in volume (V/V_0) and
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18 **the true strain in the Z-direction ϵ_{33} by the formula**
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$$\nu = -(\ln(V/V_0) - \epsilon_{33}) / 2\epsilon_{33} \quad (1)$$

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32 The use of overall volume changes rather than using x and y strains also had the benefit
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34 that small amounts of shear introduced into the cell upon stretching, caused by
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36 anisotropic cell construction, did not complicate the calculations. Typical times for a
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38 complete construction and analysis on one unit cell were about 1 week. Each mesogenic
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40 structure was repeated twice with a different starting conformation, and the average
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42 reported. The results for the Poisson's ratios on different starting conformations for the
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44 same polymer were similar within 0.1 absolute units.
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53 **Results**

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3 Two series of polymers were analysed. The first were two control polymers C5 and C7,
4 with no swivelling mesogens. They both showed a Poisson's ratio in the range of **0.5-0.6**
5 (Table 1). These are slightly higher than might be expected for commercial
6 semicrystalline or amorphous polymers (0.3-0.4) and closer to that of rubbers (0.5). It
7 may be that the high alignment is having an effect, or it may be the modelling method
8 used here. However, the Poisson's ratio of many polymers increases from around 0.3
9 under actual dynamic strain and can reach to well over 0.5 just before necking [19], and
10 these results may also be a reflection of this. In any case, this present study is initially
11 more interested in overall trends rather absolute values, especially as all the polymers
12 presented here are hypothetical at present.
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27 Typical results for the stretching dynamic simulation are shown in Figures 7 and 8. At
28 least 100K steps were needed until the volume and Z-axis settled down to a constant
29 value.
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34 The polymers S3 to S7 were constructed so that the alkyl polymer-chain length was
35 increased as the number of phenyl groups increased. This was to allow it to allow it to
36 wrap around the phenyl-based mesogen in its aligned state (figure 2). All these three
37 polymers showed much lower Poisson's ratios than the control series, and in the case of
38 S7 a clear negative value was obtained. These polymers also all showed much higher
39 strain values for the same elongational force than the equivalent control polymers. This is
40 undoubtedly because the alkyl chain is more deformed around the swivelling mesogen,
41 and as force is applied the chain is able to stretch out more easily than in the control
42 cases. In the control polymers the chain is already well aligned in the strain direction, and
43 has less free play.
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6 **The size of the both the end-to-end (C) and swivelling (S) mesogens increased in the**
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8 **series studied. To see whether the increase of the C length was having an effect on**
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10 **the Poisson's ratio as well, we looked at a polymer with a short C length of 3**
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12 **phenyls with a longer S length of 5 phenyls. As seen in Table 1, the mixed C3/S5**
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14 **polymer had a similar Poisson's ratio to the S5 polymer. The absolute value was**
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16 **slightly lower than the S5 polymer, which is probably due to the greater amount of**
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18 **swivelling mesogen per unit volume in the former. Thus the length of the in-line**
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20 **mesogen only has a small indirect effect on the Poisson's ratio, unlike that of the**
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22 **swivelling mesogen.**
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30 As predicted, the swivelling mesogens all rotated from (on average) parallel to almost
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32 exactly perpendicular to the chain direction. Preliminary modelling by us on the
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34 mesogens used by Griffin and co-workers [8, 9] (Figure 3a and b) show that they do not
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36 rotate quite as much. However, much more work needs to be done to quantify this more
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38 exactly. Nevertheless, the dialkoxydicyclobutadiene swivelling unit appears optimal
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40 based on this study. As mentioned before it is too chemically unstable to be of practical
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42 value. However similar cyclobutane or heterocyclic rings are more readily accessible, and
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44 may offer almost as good performance.
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51 It is clear from the results that to achieve a substantial lowering of the Poisson's ratio
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53 long swivelling mesogens are needed, of at least 5 rigid cyclic units. One problem in
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55 practice is that such materials with long mesogens rapidly become crystalline rather than
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3 liquid crystalline. This then causes alignment to become difficult, and the swivelling
4 motion then becomes impossible due to the “frozen-in” nature of the crystalline blocks.
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6 This might be circumvented by careful design of the mesogens to discourage
7 crystallinity. Another practical problem is that the concentration of the swivelling
8 mesogens needs to be less than that of the end-to-end mesogens in order for the director
9 to point in the chain direction rather than orthogonal to it. The lower concentration of
10 swivelling mesogens would then lower the expected auxetic response. We will report the
11 effect of concentration and linker type on the magnitude of the auxetic effect in future
12 studies.
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26 In summary, we have shown that auxetic liquid crystalline materials appear possible
27 based upon a swivelling motif. The longer the mesogenic length the lower the Poisson’s
28 ratio, with a predicted negative Poisson’s ratio somewhere between a mesogenic length
29 of between 5-7 cyclic units.
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32 List of Figures and Tables

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36 Figure 1 Reentrant rigid molecular structure (after Evans and coworkers [1])
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41 Figure 2 Liquid crystalline polymer structure containing swivelling mesogens before and
42 after stretching
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48 Figure 3 Potential linking groups in the swivelling mesogen
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53 Figure 4 Stretching of the main chain without affecting the swivelling angle
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3 Figure 5 Structures of the polymers studied, C = control mesogen, S = swivelling
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5 mesogen
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10 Figure 6 Density versus time for a dynamic simulation run of polymer S7 under a stress
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12 of 0.15 Gpa.
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17 Figure 7 Z-axis length versus time for a dynamic simulation run of polymer S7 under a
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19 stress of 0.15 Gpa.
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24 Table 1 Mechanical properties of the polymers from dynamic simulation. Each value is
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26 an average of two runs, the Poisson values for each run are calculated before averaging.
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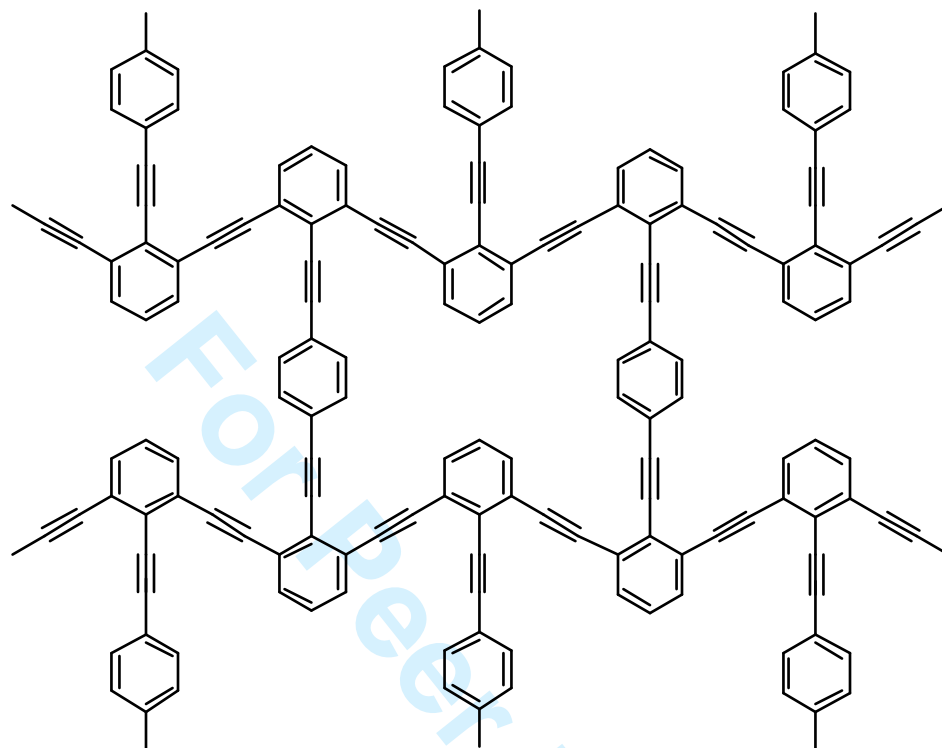


Figure 1.

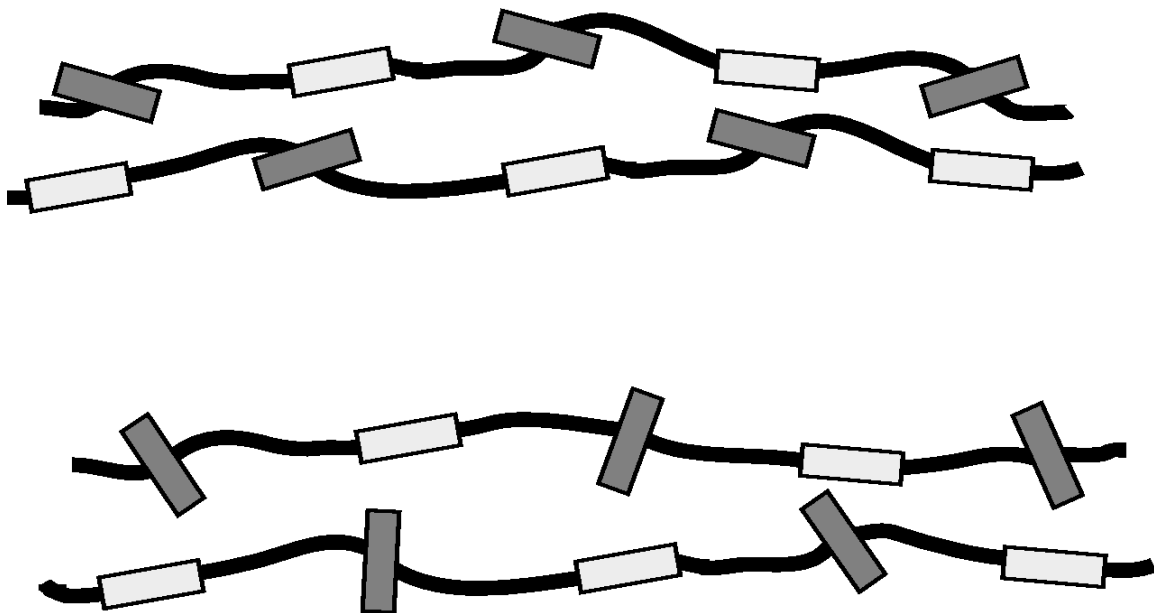


Figure 2

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Figure 3

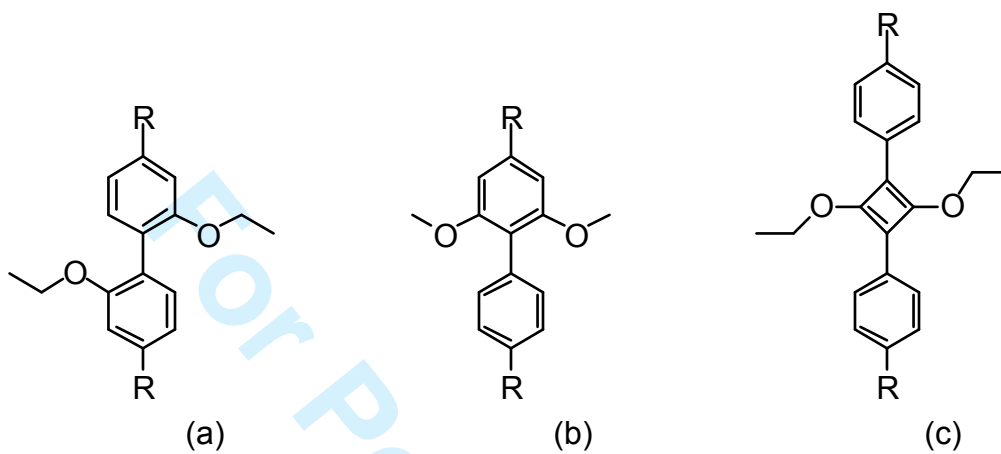
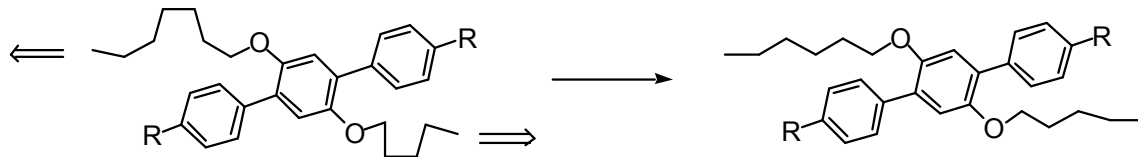
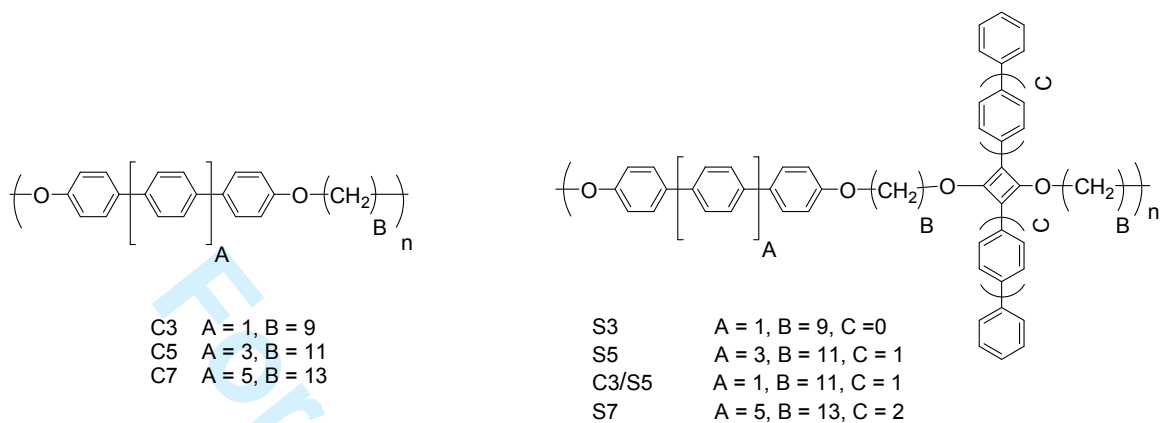


Figure 4



For Peer Review Only

Figure 5



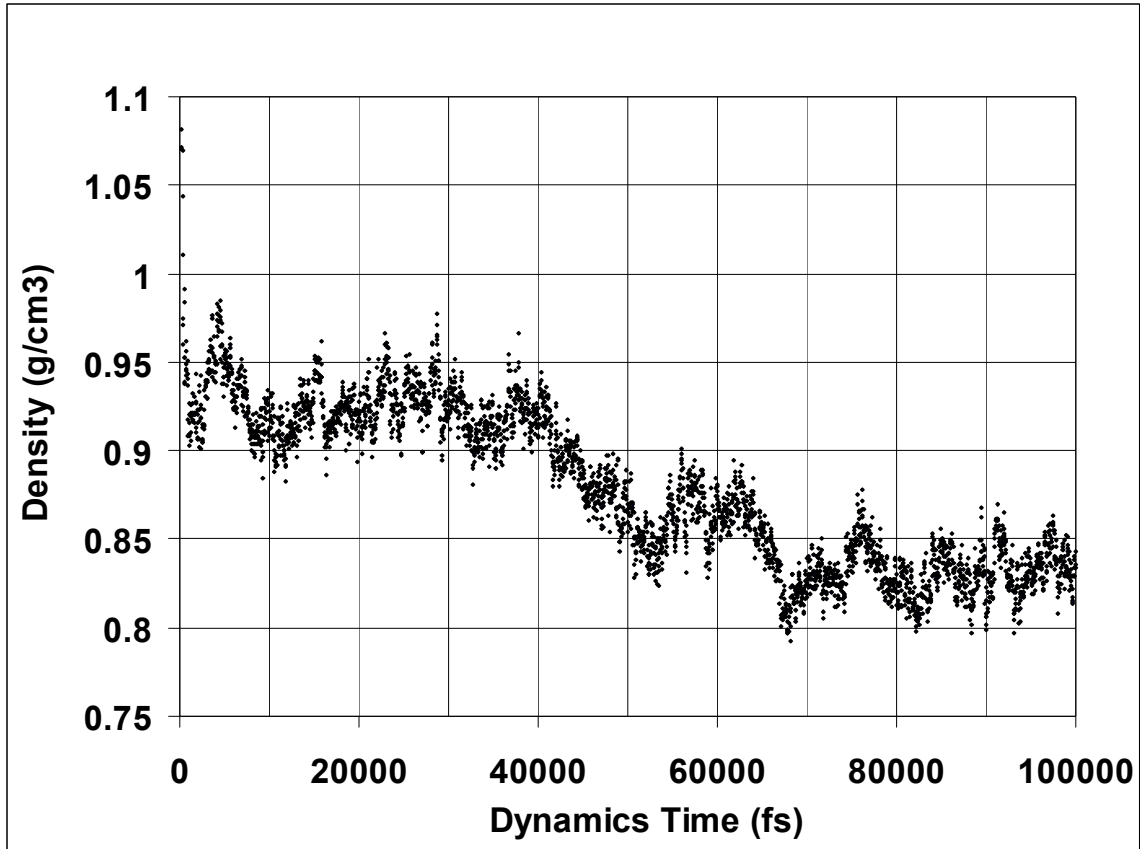
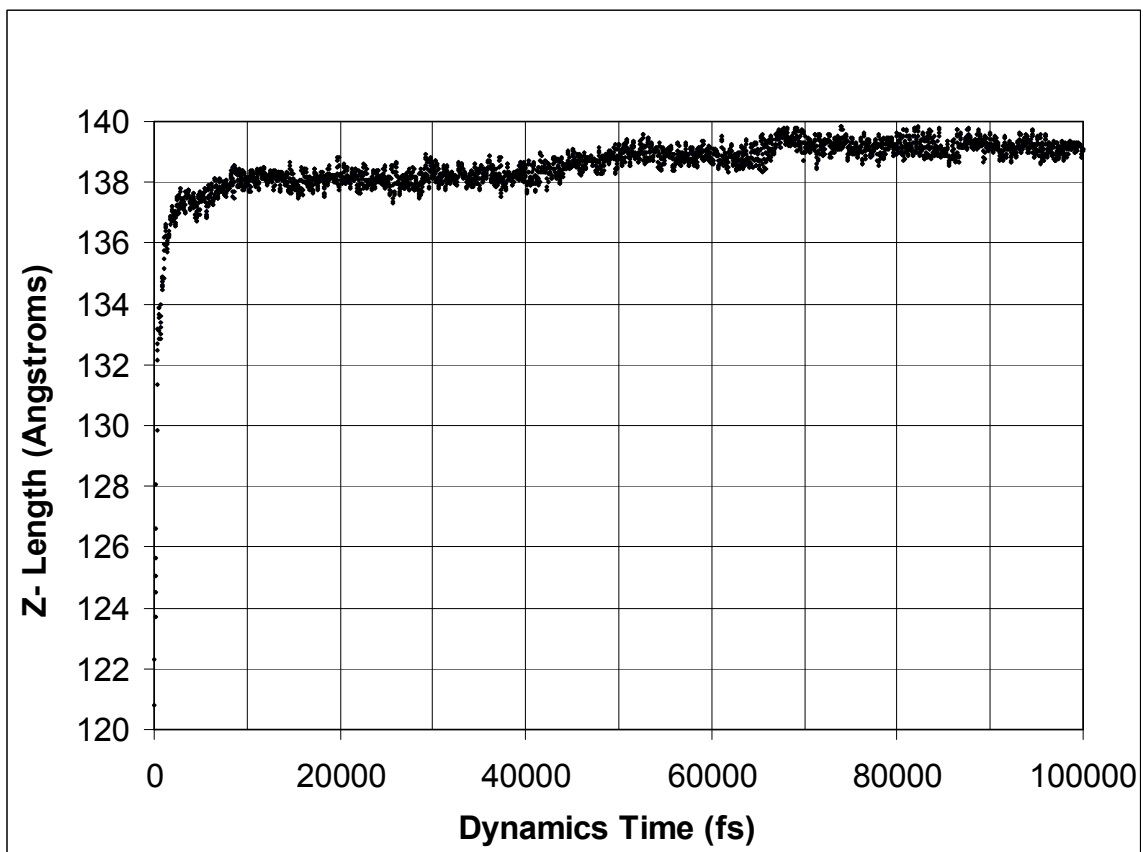


Figure 6

Review Only

Figure 7.



Polymer	Stress Applied/GPa	Final Strain ($\ln(Z/Z_0)$)	V/V _o	Poisson's ratio
C5	0.15	0.072	0.992	0.53
C7	0.1	0.048	0.99	0.59
S3	0.1	0.16	1.09	0.24
C3/S5	0.15	0.23	1.23	0.07
S5	0.15	0.25	1.20	0.15
S7	0.1	0.15	1.40	-0.60

Table 1