Treloar's Biaxial Tests and Kearsley's Bifurcation in Rubber Sheets

R.C. BATRA
Department of Engineering Science and Mehanics, Virginia Polytechnic Institute & State State University, Blacksburg, VA 24061–0291, USA

INGO MUELLER
Technical University Berlin, Limburger Strasse 20, D-1000 Berlin, Germany

PETER STREHLOW
Physikalisch-Technische Bundesanstalt Berlin, Germany

(Received 30 July 2003; accepted 3 January 2004)

Dedicated to Michael Hayes

Abstract: We review Treloar's biaxial experiments and Kearsley's calculated instability in a square rubber sheet. Treloar's rubber is recognized as a Mooney–Rivlin material. It turns out that Treloar's test could not have anticipated Kearsley's stability analysis, since his loads were far below the critical load of bifurcation. We repeat Treloar's experiment but with much higher loads and confirm Kearsley's prediction.

Key Words: rubber elasticity, bifurcation, symmetry breaking

1. RUBBER IN BIAXIAL LOADING

In the early days of non-linear elasticity, particularly for rubber, an important question concerned the validity of the kinetic theory of rubber. While that theory had not performed too badly in uniaxial stress–stretch experiments, it turned out to be qualitatively wrong for biaxial loadings.

In Treloar's time Mooney [1] had already proposed an alternative to the kinetic theory and Rivlin was working on incorporating that improvement into the emerging systematic theory of non-linear elasticity. Therefore Treloar tested what we now call the Mooney–Rivlin constitutive equation for rubber and found it satisfactory for the swollen rubber with which he experimented [2].

Years later, when Kearsley reexamined Treloar's experimental data, he detected an oddity: a square membrane dead-loaded with *equal* loads on its sides exhibited *unequal* stretches [3]. While Treloar ignored that feature, Kearsley was sensitive to it, since he, in his calculations, had detected a bifurcation which implied that equal stretches for equal loads are unstable beyond a critical value of the loads. Kearsley assumed that Treloar's data confirmed his calculation.

Mathematics and Mechanics of Solids **10:** 705–713, 2005 DOI: 10.1177/1081286505043032

f_1 (gm.)	f_2 (gm.)	λ_1	λ_2
100	100	1·08$_8$	1·08$_8$
100	200	0·99$_6$	1·30$_3$
200	200	1·21$_2$	1·19$_3$
300	200	1·55	1·06$_8$
300	100	1·66	0·88$_2$
300	300	1·41	1·36$_4$
100	400	0·80	2·04
200	400	0·95$_6$	1·95$_3$
300	400	1·24	1·83
400	400	1·65	1·65$_4$
400	500	1·50	2·12
500	500	1·97	1·94
500	300	2·31	1·10
500	200	2·43	0·85$_4$
500	100	2·48	0·73$_6$

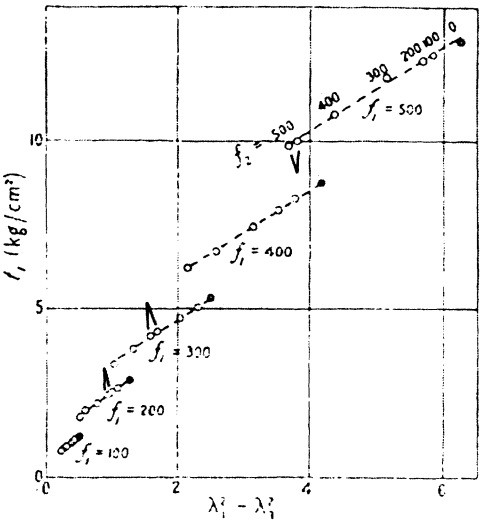

Figure 1. Treloar's data from a biaxial test and their graphical representation in a $(t_1, \lambda_1^2 - \frac{1}{\lambda_1^2\lambda_2^2})$-plot.

Unfortunately, when we check the numbers, we find that Kearsley's critical load is more than twice as high as any load ever employed by Treloar. Therefore Treloar's unequal stretches have nothing in common with Kearsley's bifurcation. Rather, the unequal stretches suggest an anisotropy which has developed in Treloar's experiments during the loading process.

Thus we concluded that Kearsley's bifurcation had never been observed and we decided to establish its existence experimentally. The experiment was successful, and its result is reported in Section 5 of this paper.

2. TRELOAR'S EXPERIMENT REVISITED

Treloar performed biaxial dead-loading tensile tests on a square sheet of swollen rubber. Those data are reproduced in Figure 1 along with Treloar's graphical representation of the measured values in a $(t_1, \lambda_1^2 - \frac{1}{\lambda_1^2\lambda_2^2})$-plot. $t_i (i = 1, 2)$ are the normal Cauchy stresses in the 1- and 2-direction, λ_i are the corresponding stretches. There is no stress in the 3-direction and the stretch λ_3 in that direction is equal to $1/\lambda_1\lambda_2$ because of incompressibility. $f_i = A_0\, t_i/\lambda_i$ are the loads in the 1- and 2-directions and A_0 is the undistorted area on which the loads act and in Treloar's experiment its value is 0.0985 cm^2.

The $(t_1, \lambda_1^2 - \frac{1}{\lambda_1^2\lambda_2^2})$ -plot was chosen by Treloar since it exhibits most clearly the deviation of the material from a neo-Hookean constitutive relation which is the one derived from the kinetic theory of rubber. That theory was fairly new – and quite popular – in 1947. The stress–stretch laws predicted by the kinetic theory read as

Table 1. Measured pairs (f_1, f_2) for a swollen rubber.

$f_2 \backslash f_1$	100	200	300	400	500
100	X		X	X	
200	X	X	X	X	
300			X	X	
400				X	X
500	X	X	X		X

$$t_1 = \left(\lambda_1^2 - \frac{1}{\lambda_1^2\lambda_2^2}\right) s_+,$$
$$t_2 = \left(\lambda_2^2 - \frac{1}{\lambda_1^2\lambda_2^2}\right) s_+, \tag{2.1}$$

so that a graph of the $(t_1, \lambda_1^2 - \frac{1}{\lambda_1^2\lambda_2^2})$-relation should be a straight line through the origin, irrespective of the load f_2. s_+ is a material constant.

What Treloar observed instead were the "sloping steps" exhibited in Figure 1 – one step for each value of f_1. It is worth mentioning that Treloar a priori assumed isotropy of his square sheet. This is why he was able to plot 25 open circles from only 15 measurements. The measurements which he did make are those for the pairs (f_1, f_2) marked in Table 1 by crosses. The unmarked pairs were filled by Treloar, via the isotropy assumption, by interchanging (f_1, f_2) and (λ_1, λ_2).

There is an oddity though in Treloar's experiments, because in his tables all equal pairs (f_1, f_2) do occur. Inspection of the table in Figure 1 shows that as a rule the measured values of λ_1 and λ_2 are unequal when the loads f_1, f_2 are equal – a glaring sign of anisotropy which Treloar does not comment upon. What he does is draw *two* circles for the pairs of equal forces: one circle for the measured values of (λ_1, λ_2) and the other one for those stretches interchanged. In this manner Treloar comes to 30 circles. Sometimes the circles for equal forces overlap but often they do not. In Figure 1 we have identified the double occurrences by the chevrons in Treloar's plot, wherever they are clearly visible.

In addition Treloar introduces black dots into Figure 1 for the result of uniaxial loading tests in the 1-direction. He does not give values for those.

3. CONFIRMATION OF THE MOONEY–RIVLIN RELATION

Having convincingly established that the neo-Hookean constitutive relation is invalid for his rubber Treloar proceeds to test an alternative. That alternative is due to Mooney and is now known as the Mooney–Rivlin constitutive relation. For biaxial loading of the Treloar type the relation consists of two equations, namely

$$t_1 = \left(\lambda_1^2 - \frac{1}{\lambda_1^2\lambda_2^2}\right)(s_+ - s_-\lambda_2^2)$$

$$t_2 = \left(\lambda_2^2 - \frac{1}{\lambda_2^2\lambda_1^2}\right)\left(s_+ - s_-\lambda_1^2\right), \tag{3.1}$$

where $s_\pm$ are constants, dependent on material.

Table 2. $s_\pm$ according to Treloar's measurements for swollen rubber.

	100	300	300	100	200	300	400	500	500	500
	200	200	100	400	400	400	500	300	200	100
$s_-[\mathrm{g/cm^2}]$	-200	-397	-337	-250	-292	-251	-271	-221	-245	-168
$s_+[\mathrm{g/cm^2}]$	2186	1884	1946	2028	1981	1669	2059	1996	1993	2061

Treloar's data may be used to obtain $s_\pm$ by solving the two equations (3.1) for $s_\pm$. We have

$$\begin{aligned} -s_- &= \frac{1}{A_0}\frac{1}{\lambda_2^2 - \lambda_1^2}\left(\frac{f_1\lambda_1}{\lambda_1^2 - \frac{1}{\lambda_1^2\lambda_2^2}} - \frac{f_2\lambda_2}{\lambda_2^2 - \frac{1}{\lambda_2^2\lambda_1^2}}\right), \\ s_+ &= +s_-\lambda_2^2 + \frac{1}{A_0}\frac{f_1\lambda_1}{\lambda_1^2 - \frac{1}{\lambda_1^2\lambda_2^2}}. \end{aligned} \tag{3.2}$$

We take the data from Figure 1 and obtain values for $s_\pm$ which are listed in Table 2, where the first double line indicates the loads $f_{1,2}$ – in grams (!); we stick to Treloar's somewhat archaic units while discussing his experiment.

The scatter is wild indeed, particularly in s_-. If we take average values, we obtain

$$s_- = -251\frac{\mathrm{g}}{\mathrm{cm}^2} \qquad \text{and} \qquad s_+ = 2010\frac{\mathrm{g}}{\mathrm{cm}^2}. \tag{3.3}$$

With these average values inserted into (3.1) we may recalculate, largely numerically, the position of Treloar's circles in Figure 1_{right} according to the Mooney–Rivlin constitutive equation. Figure 2_{left} , shows the result of that calculation. Tht line in the dotted straighe figure – marked by asterisks – corresponds to uniaxial tension in the kinetic theory. It follows that the kinetic theory is not too bad for uniaxial stress–stretch experiments. Comparison of Figure 2_{left} with the experimental plot in Figure 1 shows good agreement. Thus Treloar was able to say: "Detailed comparison shows a very close quantitative correspondence between the theoretical and the experimental points". In this manner Treloar firmly established the validity and usefulness of the Mooney–Rivlin stress–stretch relation.

For good measure we also plot t_1 vs. $\left(\lambda_1^2 - \frac{1}{\lambda_1^2\lambda_2^2}\right)\left(s_+ - s_-\,\lambda_2^2\right)$ from Treloar's experimental data with $s_\pm$ as in (3.3). Thus we obtain an essentially straight graph equal to the bisector, cf. Figure 2_{right} , as it must be according to the Mooney–Rivlin law.[1]

Treloar also presents experimental data for dry rubber and he is less happy with those as regards to the Mooney–Rivlin theory. Indeed, Treloar's dry rubber departs significantly from that theory.

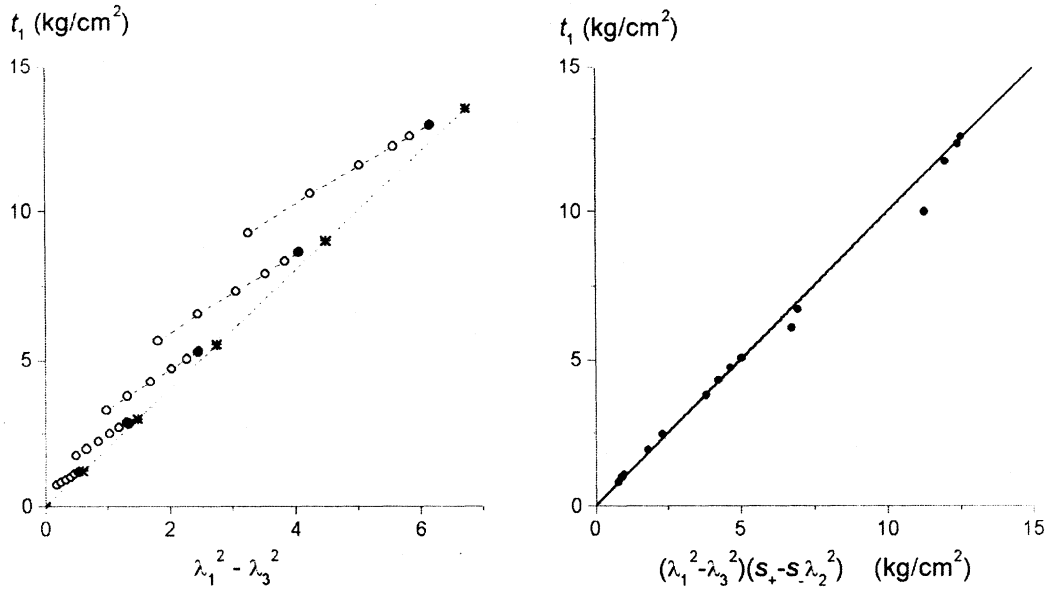

Figure 2. Left: Treloar plot for Mooney–Rivlin theory with coefficients from (3.3). Right: t_1 vs. $\left(\lambda_1^2 - \frac{1}{\lambda_1^2\lambda_2^2}\right)(s_+ - s_-\lambda_2^2)$ from Treloar's data (dots) and from Mooney–Rivlin theory (straight line).

All this is old history revisited. The motivation for bringing this up comes from a suggestion made by Kearsley [3] in a more recent paper on the stability of a biaxially loaded square sheet of Mooney–Rivlin rubber. Kearsley sees some of this theoretical findings confirm Treloar's old experimental results. We proceed to explain.

4. KEARSLEY'S INSTABILITY

Kearsley made a mathematical study of a square Mooney–Rivlin membrane subject to equal dead loads in the direction of the edges. From (3.1) with $f_i = A_0 t_i/\lambda_i$ he concluded that under those circumstances λ_1 and λ_2 must satisfy the condition

$$(\lambda_1 - \lambda_2)\left\{\left(\lambda_1^3\lambda_2^3 + 1\right)\frac{s_+}{|s_-|} + \left(\lambda_1^2 + \lambda_1\lambda_2 + \lambda_2^2 - \lambda_1^4\lambda_2^4\right)\right\} = 0. \tag{4.1}$$

One solution is obviously $\lambda_1 = \lambda_2$, i.e. symmetric stretch under symmetric loading and that is the solution which is intuitively expected. But (4.1) can also be satisfied by the curly bracket term being equal to zero. This solution is non-symmetric and depends on the coefficient $\frac{s_+}{|s_-|}$ which in Treloar's case, by (3.3), has the value 8. Figure 3 shows the two solutions in a (λ_1, λ_2)-plot. The point of intersection of bifurcation occurs at $\lambda_1 = \lambda_2 = \lambda_f$ with

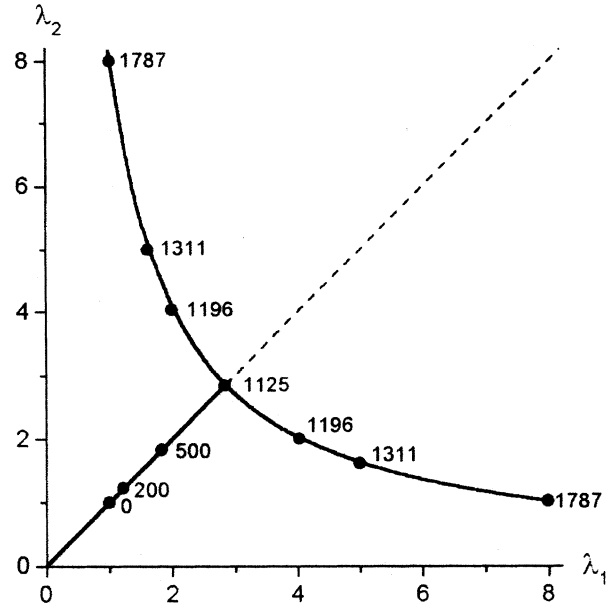

Figure 3. Bifurcation in the (λ_1, λ_2)-diagram. Numbers indicate the values of $f_1 = f_2$ in g.

$$\left(1+\lambda_f^6\right)\frac{s_+}{|s_-|}+\lambda_f^2\left(3-\lambda_f^6\right)=0, \quad \text{hence} \quad \lambda_f = 2.84. \tag{4.2}$$

The forces corresponding to the bifurcation point are accordingly

$$f_1 = f_2 = A_0\,|s_-|\left(\lambda_f - \frac{1}{\lambda_f^5}\right)\left(\frac{s_+}{|s_-|}+\lambda_f^2\right) = 1125\ \text{g}. \tag{4.3}$$

Moreover, Kearsley proved that the symmetric solution $\lambda_1 = \lambda_2$ is unstable beyond the bifurcation point. Therefore for loads $f_1 = f_2$ bigger than 1125 g we expect to see different stretches λ_1 and λ_2, one bigger than $\lambda = 2.84$ and the other one smaller.

This fact gave Kearsley an idea: had not Treloar observed different stretches for equal loads? Indeed he had, and he had put them down in his table and diagram, cf. Figure 1. Therefore Kearsley, apparently without looking at the numbers, announced that there was "an experimental example of this sort of asymmetric solution to a dead-loading problem...".

This interpretation of Treloar's results is wrong. Indeed, according to Figure 3 we do not expect unequal data for λ_1 and λ_2 except for loads beyond $f_1 = f_2 = 1125$ g. and then one of the stretches must be bigger than 2.84. But Treloar's highest equal loads were 500 g according to Figure 1 and the biggest stretch in his table was 2.48.

Therefore Treloar's data do not offer experimental evidence of Kearsley's instability. To our knowledge this instability has not been demonstrated to this day.

We must conclude, rather prosaically, that Treloar's observation of two different stretches for equal loads is nothing else but another example of a notorious quality of rubber, namely the

difficulty of quantitative reproducibility of rubber data and the unreliability of exact numbers obtained from rubber experiments.

5. AN EXPERIMENT

And yet, it would clearly be desirable to prove Kearsley's bifurcation experimentally. Therefore we chose a square membrane of rubber of area 5 cm×5 cm, and undistorted thickness of 0.4 mm, cf. Figure 4, top. We stretched this membrane by the application of equal dead loads.

The square membrane was cut from the flat part of a commercially available balloon made from weakly vulcanized natural rubber. Its disk-like shape of diameter 12 cm in the undistorted state is changed into a perfectly spherical form by inflating the balloon . Before the square piece was cut out, the balloon was inflated in order to record its pressure-radius curve by means of a pressure gauge. Fitting this curve to the pressure-radius relation of a Mooney–Rivlin balloon provided a pair of values for the elastic constants $s_{\pm}$. More values for $s_{\pm}$ were obtained during the biaxial loading of the membrane from six stretch measurement, at six different loads. The loads were applied quasistatically and the resulting stretches were read off some minutes after the application of loads. Thus it was determined that the coefficients for the rubber membrane had the values

$$s_+ = 1.858 \times 10^5 \text{ Pa}, \qquad s_- = -0.1935 \times 10^5 \text{ Pa},$$

with a relative standard uncertainty of 7×10^{-2}.

Therefore the bifurcation stretches should be equal to $\lambda_1 = \lambda_2 = 3.106$ and the critical loads should have the values $f_1 = f_2 = 23.12$ N.

At this level of the loads the membrane turned out to be quite "soft", i.e. a slight touch of one set of weights or the other could deform the membrane into either one of the two possible rectangles. This is typical for a bifurcation. After a slight increase of the load to 23.3 N in both directions we observed that the original square of 1 cm×1 cm in the center of the membrane was deformed to the rectangular area of size 3.05 cm×4.06 cm, cf. Figure 4, bottom.

Thus we concluded that our specimen exhibits the Kearsley bifurcation, and that the load 23.3 N was supercritical as expected.

We have calculated the available free energy function of the membrane under equal loads and plotted its contour lines for two sets of loads, one subcritical and the other one supercritical. Figure 5 shows these plots which are taken from the book on Rubber and Rubber Balloons [4]. The left figure shows the energetic minimum in a symmetric position, while in the right figure that symmetric position is occupied by a saddle. The symmetry has been broken and there are now two minima. Actually the landscape around these minima and the saddle is quite "flat" which accounts for the softness of the membrane at and around the bifurcation point.

After unloading the membrane exhibited slight residual stretches not unlike those of a deflated rubber balloon. Those stretches disappeared during gentle heating and the membrane could be used, and has been used repeatedly, in further experiments of the same type.

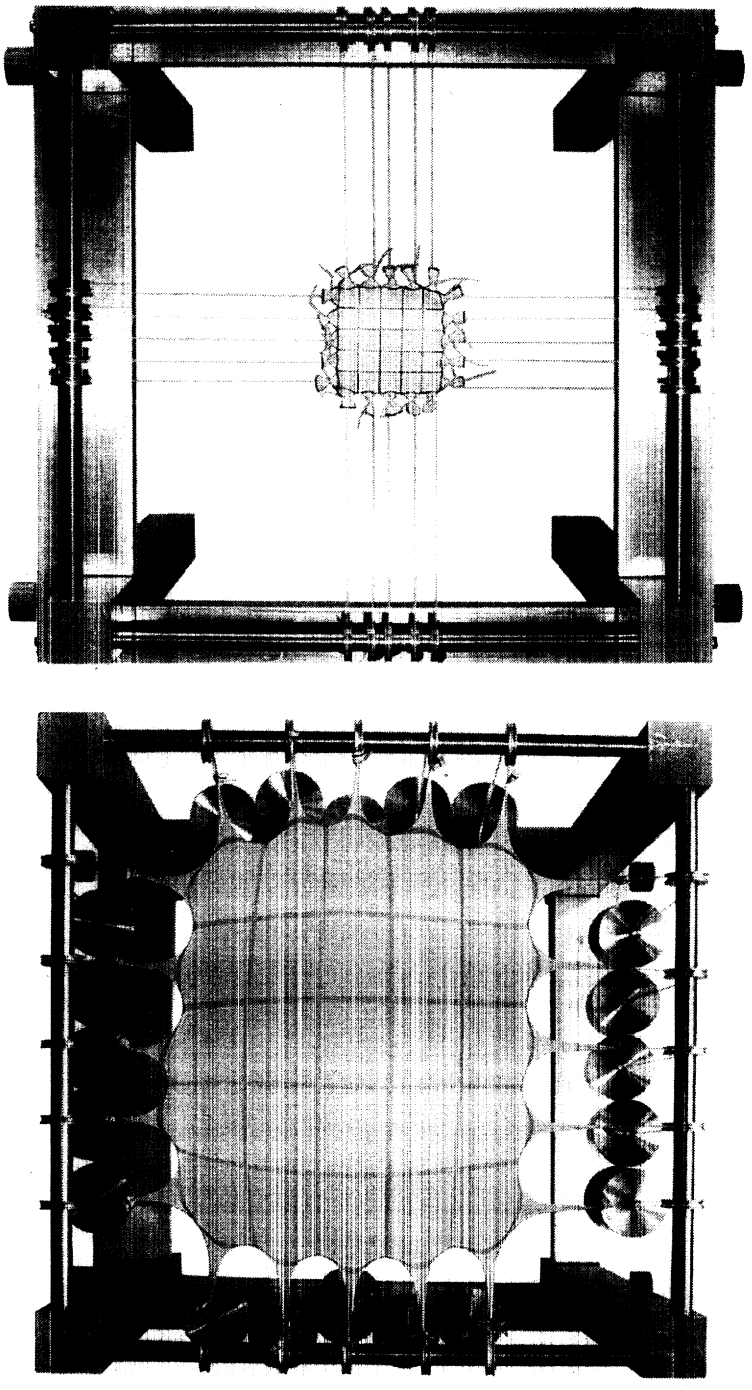

Figure 4. Instability of a square rubber membrane under equal loads (photographs). Top: Undistorted state Bottom: Supercritical loads.

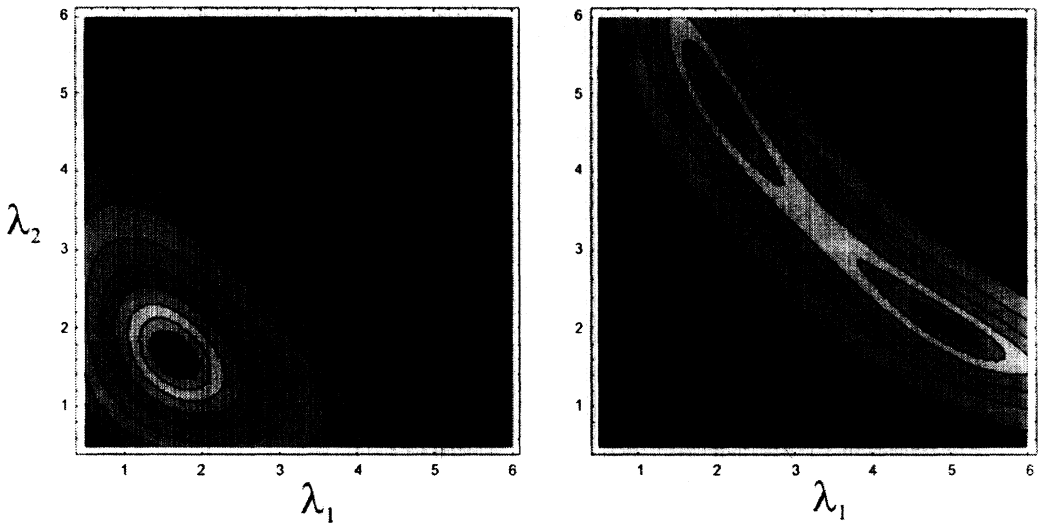

Figure 5. Contour lines of available free energy for a sub-critical load (left) and a supercritical one (right).

No inhomogeneity could be detected in our sample even after loading and no loss of isotropy could be noticed after unloading and gentle heating.

NOTE

1. Treloar draws a similar graph for $\frac{|s_-|}{s_+} = 0.1$ and also confirms the Mooney–Rivlin law. Since our $\frac{|s_-|}{s_+}$ equals 0.125, it seems that the exact value of s_- is not overly important in this experiment.

REFERENCES

[1] Mooney, M. A. Theory of large elastic deformation. *Journal of Applied Physics,* 11, 582–592 (1940).

[2] Treloar, L. R. G. Stresses and birefringence in rubber subjected to general homogeneous strain. *Proceedings of the Physical Society,* 60, 135–145 (1947).

[3] Kearsley, E. A. Asymmetric stretching of a symmetrically loaded elastic sheet. *International Journal of Solids and Structures,* 22, 111–119 (1985).

[4] Müller I. and Strehlow, P. *Rubber and Rubber Balloons.* Lecture Notes on Physics, Springer, Berlin (in press).