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Note on the time-dependent deformation of a viscous drop which is almost spherical

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The theory of the shear-induced small deformation of viscous drops at zero Reynolds number is reviewed. The general result for arbitrary shear and surface tension is presented, and the asymptotic forms for weak flow and for high internal viscosity are derived from it. In the latter case, numerical solutions are compared with the experiments of Torza, Cox & Mason (1972).

1. Introduction

A number of papers, starting with the pioneering work of Taylor (1934) have dealt with the theory of deformation of viscous drops. Many of these (Taylor 1934; Cox 1969; Frankel & Acrivos 1970; Barthès-Biesel & Acrivos 1973*a*, *b*) have concerned deviations from sphericity which are small. There are, however, discrepancies between the various papers, and in consequence some confusion has arisen as to which of the proposed equations is appropriate to describe experimental results – such as those obtained by Hakimi & Schowalter (1980). The purpose of this note, then, is to unify the theoretical results, and to make clear the conditions under which each applies, together with its order of accuracy.

In conformity with the notation of Hakimi & Schowalter (1980) we consider an incompressible drop of viscosity μ^* and volume $\frac{4}{3}\pi b^3$ immersed in an unbounded fluid of viscosity μ_0. The suspending fluid is sheared at a rate $G(\mathbf{E}+\mathbf{\Omega})$ with $\mathbf{E}$ and $\mathbf{\Omega}$ the symmetric and antisymmetric parts of the velocity gradient, and the consequent drop deformation is inhibited by the interfacial surface tension σ. We suppose, as in the papers quoted above, that the deviation from sphericity is small.

The feature which all the analyses have in common and attempt to exploit is that for a nearly spherical drop the flow field may be determined by a perturbation expansion and, due to the quasi-stationarity implied by the Stokes equations, is determined by the instantaneous shape. Since in general the deformation becomes not small eventually, the perturbation scheme by which the velocity field is generated cannot be justified for all times. The time for which the analysis does remain valid varies according to the physics in each case as we show below.

In a time-dependent problem, then, we may legitimately postulate a near-sphere initial condition for the drop shape, and the natural questions to be asked are, first, which terms are important in the shape evolution equation; and, secondly, for how long the assumption of near-sphericity remains valid.

Given some shape S for the drop whose deviation from sphericity is measured by the

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small parameter ϵ, we suppose that the solution $\mathbf{u}(\mathbf{x})$ of the Stokes equations for the fluid velocity everywhere is known. By linearity, this consists of the sum of two terms, one linear in the forcing G by the external flow, and the second proportional to the surface tension σ of the drop. The latter vanishes with ϵ, and so we may write

$$\mathbf{u} = bG\mathbf{u}_1 + \frac{\epsilon\sigma}{\mu_0}\mathbf{u}_2,$$

with each $\mathbf{u}_i = \mathbf{u}_i(\mathbf{x}, \mu^*/\mu_0, \epsilon, S)$ and $\mathbf{u}_1$ depending also on the tensorial nature of the applied linear flow. Now non-dimensionalizing time with respect to the shear rate G^{-1}, we have for the normal velocity at the drop surface

$$\mathbf{u}.\mathbf{n} = u_{1n} + k\epsilon u_{2n}, \tag{1}$$

where, as in Hakimi & Schowalter (1980), we write $k = \sigma/bG\mu_0$ and $\lambda = \mu^*/\mu_0$. It is, of course, only the normal velocity that is responsible for the time-dependent drop deformation. We see at once from (1) that, if u_{1n}/k is not $O(\epsilon)$ or smaller, then the deformation will not remain small after a time of order unity (or of order λ for large λ), and hence the solution will break down. We consider below the special cases in which u_{1n}/k is small, but consider the general case first.

In principle, this approach demands that we consider a surface of arbitrary shape (provided its deviation from sphericity is small) and consider the excitation and relaxation of *all* possible modes of distortion. This has been done by Cox (1969) but only for $\lambda = \infty$. The results are rather complicated. In addition, we see that, if the deviation from sphericity is measured by spherical harmonics of all orders, then since the flow itself is second harmonic in type, at leading order in ϵ the rate of change of the nth harmonic is, at worst, forced only by harmonics of orders $n, n+2$. In consequence if the initial condition for the shape is spherical or specified by a second harmonic (and these are the common cases of interest) then at leading order in ϵ the subsequent deformation may be described by a second harmonic, although higher accuracy in ϵ requires correspondingly higher harmonics.

We consider then the instantaneous surface

$$r = b(1+\epsilon f) \quad \text{with} \quad f = r^3\mathbf{F}:\nabla\nabla(1/r) + \epsilon[(-6/5)\,\mathbf{F}:\mathbf{F} + r^5\mathbf{H}\,\vdots\,\nabla\nabla\nabla\nabla(1/r)] \tag{2}$$

in which $\mathbf{F}$ and $\mathbf{H}$ are completely symmetric and traceless tensors of the second and fourth ranks, respectively, and for which the normal surface velocities u_{1n}, u_{2n} have been determined by Barthès-Biesel & Acrivos (1973*a*). As noted above, we may expect that the flow, although it will not preserve the shape defined by (2) as the deformation proceeds, will generate higher harmonics in f only at higher orders in ϵ. We therefore have, slightly modifying Barthès-Biesel & Acrivos' analysis, that

$$\epsilon\frac{\mathscr{D}\mathbf{F}}{\mathscr{D}t} = a_0\,\mathbf{E} + \epsilon\{ka_1\,\mathbf{F} + a_2\,Sd(\mathbf{E}.\mathbf{F})\} + \epsilon^2\{ka_3\,Sd(\mathbf{F}^2) + a_4\,\mathbf{EF}:\mathbf{F} + a_5\,\mathbf{FE}:\mathbf{F} + a_7\,Sd(\mathbf{E}.\mathbf{F}^2) + a_8\,\mathbf{H}:\mathbf{E}\} + O(\epsilon^3, k\epsilon^3), \tag{3}$$

$$\epsilon\frac{\mathscr{D}\mathbf{H}}{\mathscr{D}t} = b_1\,Sd_4(\mathbf{EF}) + \epsilon\{b_0 k\mathbf{H} + b_2 kSd_4(\mathbf{FF}) + b_3\,Sd_4(\mathbf{E}.\mathbf{H}) + b_4\,Sd_4(\mathbf{E}.\mathbf{FF})\} + O(\epsilon^2, k\epsilon^2), \tag{4}$$

in which $\mathscr{D}/\mathscr{D}t$ is a Jaumann derivative rotating with the vorticity $\mathbf{\Omega}$, and the 'symmetric deviator' operators Sd, Sd_4 are defined by Barthès-Biesel & Acrivos (1973*a*).

They also give a_i ($i = 0, 8$); b_i ($i = 0, 2$) explicitly as (rational) functions of λ. In particular, $a_0 = 5/(2\lambda+3)$. The coefficients b_3, b_4 are similarly functions of λ but are not known. We expect that, for all values of λ, a_i, b_i (and corresponding coefficients for higher-order terms) will be no larger than $O(1)$ but the limit $\lambda \to \infty$ is of particular concern. We shall see below that the boundedness requirement *is* met in the high λ limit, and, on the assumption that the velocities vary continuously with λ, it follows that the coefficients are bounded for all values of λ. The equations above differ from those of Barthès-Biesel & Acrivos in two respects. First, we have made no assumption about the magnitude of k (whereas they take $k = \epsilon^{-1}$). Secondly, certain higher-order terms (e.g. $\epsilon^3 a_6 k\mathbf{F}:\mathbf{FF}$) are here neglected (as they may be to the order of accuracy presented).

We see from (3) that $\mathbf{F}$ becomes $> O(1)$ after a time $O(\epsilon)$ unless either k or λ is large. Hence in general the equations above accurately describe the time evolution (say from sphericity $\epsilon\mathbf{F} = \epsilon^2\mathbf{H} = 0$) only for small times, and no equilibrium is possible within the range of validity of the analysis. In addition, while the analysis remains valid it may be seen that, for $O(\epsilon)$ accuracy in the shape, the $O(\epsilon^2)$ terms in (3) can be neglected, and (4) ignored completely. Similarly at $O(\epsilon^2)$ (3) and (4) suffice: the sixth harmonic terms can be neglected and so the initial shape postulated in (2) is preserved with $\mathbf{F}$ and $\mathbf{H}$ regarded as functions of time, and errors in r of $O(\epsilon^3)$.

We now turn to the special cases in which an equilibrium does exist within the range of validity of the analysis.

2. Weak flow: $k \gg 1$ with $\lambda = o(k)$

Since in principle k, ϵ are independent parameters for the system (3), (4) may be simplified for the effect of $k \gg 1$ without reference to ϵ. On the other hand if the weak flow is assumed to be responsible for the small deviation from sphericity then we may put $k = \epsilon^{-1}$. This is the case we consider. Thus (3), (4) become

$$\epsilon \frac{\mathscr{D}\mathbf{F}}{\mathscr{D}t} = a_0 \mathbf{E} + a_1 \mathbf{F} + \epsilon[a_2 Sd(\mathbf{E}\,.\,\mathbf{F}) + a_3 Sd(\mathbf{F}^2)] + O(\epsilon^2), \tag{5}$$

$$\epsilon \frac{\mathscr{D}\mathbf{H}}{\mathscr{D}t} = b_1 Sd_4(\mathbf{EF}) + b_0 \mathbf{H} + b_2 Sd_4(\mathbf{FF}) + O(\epsilon). \tag{6}$$

These are the equations of the '$O(\epsilon)$ theory' of Barthès-Biesel & Acrivos (1973*a*) except that the time derivative on the left-hand side of (6) is replaced by $\partial/\partial t$, as it may be to this level of approximation. They, in fact, give some of the higher-order terms but not to any consistently higher order. The $O(1)$ theory of Taylor (1934) corresponds simply to

$$\epsilon \frac{\partial \mathbf{F}}{\partial t} = a_0 \mathbf{E} + a_1 \mathbf{F} + O(\epsilon). \tag{7}$$

Only in this case it is permissible to replace the Jaumann derivative by an Eulerian time derivative. The numerical computations of Hakimi & Schowalter (1980) suggest, however, that this is a poor approximation for small but finite ϵ and for that reason we have retained also $\mathscr{D}/\mathscr{D}t$ in (6). We note in passing that the appearance of $\epsilon\,\partial/\partial t$ in (7) shows that for weak flows the appropriate time scale is the surface-tension time $\sigma/\mu_0 b(1+\lambda)$ rather than the shear time G^{-1}.

3. High viscosity drops: $\lambda \gg 1$ with $k = o(\lambda)$

It is in this case that the greatest confusion has arisen. As before, ϵ and $1/\lambda$ may be assumed independent, but the case of interest is where an $O(1/\lambda)$ distortion has been produced by the flow. We therefore put $\lambda = \epsilon^{-1}$ and simplify (3), (4). Now as shown by Barthès-Biesel & Acrivos (1973*a*), as $\lambda \rightarrow \infty$, $a_i = O(1/\lambda)$ for $i = 0, 1, 2, 3, 8$, but remains $O(1)$ for $i = 4, 5, 7$. Thus some terms 'jump order' in (3) and give

$$\frac{\mathscr{D}\mathbf{F}}{\mathscr{D}t} = \tfrac{5}{6}\mathbf{E} + \epsilon\{-\tfrac{20}{19}k\mathbf{F} + \tfrac{10}{7}Sd(\mathbf{E}\,.\,\mathbf{F}) - \tfrac{5}{4}\mathbf{E} - 3\mathbf{EF}\colon\mathbf{F} + 18Sd(\mathbf{E}\,.\,\mathbf{F}^2) - 6\mathbf{FE}\colon\mathbf{F}\} + O(\epsilon^2, k\epsilon^2), \tag{8}$$

$$\frac{\mathscr{D}\mathbf{H}}{\mathscr{D}t} = \tfrac{1}{14}\,Sd_4(\mathbf{EF}) + b_3'\,Sd_4(\mathbf{E}\,.\mathbf{H}) + b_4'\,Sd_4(\mathbf{E}\,.\,\mathbf{FF}) + O(\epsilon, k\epsilon), \tag{9}$$

where $b'_{3,4} = \lim\limits_{\lambda\rightarrow\infty} b_{3,4}$. The $O(1)$ version of these equations is simply

$$\frac{\mathscr{D}\mathbf{F}}{\mathscr{D}t} = \tfrac{5}{6}\mathbf{E}. \tag{10}$$

An improved form of (8), (9) may be obtained by rederiving (3), (4) from first principles in the limit $\lambda \rightarrow \infty$. This provides an independent check of (8), explains the jumping of orders in $1/\lambda$, and ensures that the $a_i(\lambda)$ do not exceed $O(1)$ as $\lambda \rightarrow \infty$.

The crucial idea is that at leading order in $1/\lambda$ the drop is a rigid body, which can rotate, but not deform. Only at the next order does the unbalanced external tangential stress on the surface drive a deformation. In consequence, whatever the instantaneous shape, the flows inside ($\mathbf{u}^*$) and outside ($\mathbf{u}$) the drop are given by:

$$\mathbf{u}^* = \begin{pmatrix} \mathbf{u}_0^*: \\ \text{Solid body rotation of} \\ \text{rigid body due to } G \text{ at } \infty. \end{pmatrix} + \frac{1}{\lambda}\begin{pmatrix} \mathbf{u}_1^*: \\ \text{Flow due to surface tension} \\ +\text{flow due to stress generated} \\ \text{by } \mathbf{u}_0 \end{pmatrix} + \dots,$$

$$\mathbf{u} = \begin{pmatrix} \mathbf{u}_0: \\ \text{Flow outside rigid body} \\ \text{with } G \text{ at } \infty. \end{pmatrix} + \frac{1}{\lambda}\begin{pmatrix} \mathbf{u}_1: \\ \text{Flow generated by matching} \\ \text{with}{}_1^* \end{pmatrix} + \dots.$$

At each stage, the internal velocity fields may be computed to within a rigid body motion from the known surface stresses generated at lower order (at the first stage these are zero). This then enables the outer flow, $\mathbf{u}$, to be calculated at the same order, to within that generated by the unknown rigid body motion. This motion is finally determined by the requirement that the drop exert no net force or couple on the outer fluid (the dotted arrows above).

For the surface given by (2) then, $\mathbf{u}_0^*$ is the rotation of a rigid, couple-free near-sphere in a straining motion $G(\mathbf{E} + \mathbf{\Omega})$. As noted by Rallison (1978, §7) this angular velocity $G\mathbf{\Omega}^*$ is given by

$$\mathbf{\Omega}^* = \mathbf{\Omega} - 3\epsilon(\mathbf{F}\,.\,\mathbf{E} - \mathbf{E}\,.\,\mathbf{F}) + O(\epsilon^2).$$

Hence denoting the modified Jaumann derivative with $\mathbf{\Omega}$ replaced by $\mathbf{\Omega}^*$ as $\mathscr{D}^*/\mathscr{D}t$, we have formally that

$$\mathscr{D}^*S/\mathscr{D}t = (1/\lambda)\,(\text{stretching terms: } \mathbf{u}_1, \mathbf{u}_0^*) + O(1/\lambda^2, k/\lambda^2),$$

and so with $\epsilon = \lambda^{-1}$ the $O(\epsilon)$ terms in (3), (4) give the stretching terms to sufficient accuracy, and thus

$$\mathscr{D}^*\mathbf{F}/\mathscr{D}t = \tfrac{5}{6}\mathbf{E} + \frac{1}{\lambda}[-\tfrac{20}{19}k\mathbf{F} + \tfrac{10}{7}Sd(\mathbf{E}\,.\,\mathbf{F}) - \tfrac{5}{4}\mathbf{E}] + O\left(\frac{1}{\lambda^2}, \frac{k}{\lambda^2}\right), \qquad (8)'$$

$$\mathscr{D}^*\mathbf{H}/\mathscr{D}t = \tfrac{1}{14}Sd_4(\mathbf{EF}) + O(1/\lambda, k/\lambda), \qquad (9)'$$

and these can be shown to be equivalent to (8), (9) with $b_3' = b_4' = 0$. The terms which 'jump order', then, are precisely those corresponding to a pure rotation of the near-sphere without deformation. By an extension of the same argument, we can see that those a_i's corresponding to surface deformation are no larger than $O(1/\lambda)$, and that those corresponding to rotation are $O(1)$ as $\lambda \to \infty$.

In all previous analyses of this problem, it seems that only (8) has been written down, although (9) is required at the same order of accuracy for the shape. The $O(1)$ equation (10) has been given correctly by several authors (Taylor 1934; Cox 1969). Its solutions are of two types: if the vorticity is smaller than $O(\lambda^{-1})$, then the drop deforms monotonically and the deformation becomes large after a finite time (of order λ); if the vorticity is not small, then (10) gives a regular oscillation of shape with no tendency toward equilibrium even for $t \to \infty$ (surface tension does not appear at this order, so the absence of a trend toward equilibrium is not unreasonable in view of the reversibility of Stokes flow). Thus in order to predict such an equilibrium the next terms are required. Of the terms which appear at the next order, Taylor (1934) and Cox (1969) gave the first in braces in (8); Frankel & Acrivos (1970) the first two; Barthès-Biesel & Acrivos (1973*b*) gave all, but with a typographical error [their θ_6 should be

$$\tfrac{5}{6} - \tfrac{5}{4}\lambda^{-1} - 3\lambda^{-1}A_{lm}A_{lm}$$

following their equation (18)].

Cox (1969) demonstrated, using his truncated form of (8), that the shape approached equilibrium at large times via a spiral. The corrected version shows the same qualitative behaviour as seen in figure 1, where the theoretical results here are compared with the experiments of Torza, Cox & Mason (1972). The flow considered is a simple shear with $(\mathbf{E}+\mathbf{\Omega})_{12} = 1$, and other components zero. A scalar measure of the distortion is given by $D(t) = (l-b)/(l+b)$, where l, b are the major and minor axes of the 1, 2 cross-section; and its orientation is given by $\alpha(t)$, the angle between the major axis and the 2-direction. The agreement is reasonable, with the magnitude of the distortion ($\approx 0{\cdot}1$) underestimated by 10 % and the frequency of oscillation by 5 %. A higher-order theory seems to be necessary to resolve these discrepancies.

In the case where surface tension is absent altogether ($k = 0$) no equilibrium exists and the solution still shows a periodic cycle. Figure 2 shows a polar plot of D against α for a single oscillation, which then repeats indefinitely. The experimental result for the distortion is seen to be almost twice as large as that given by (8), though the period is accurate to within 10 %. The principal reason for the discrepancy here is probably the largeness of the (transient) distortion ($\approx 0{\cdot}2$) and so the $O(\epsilon)$ theory is inadequate.

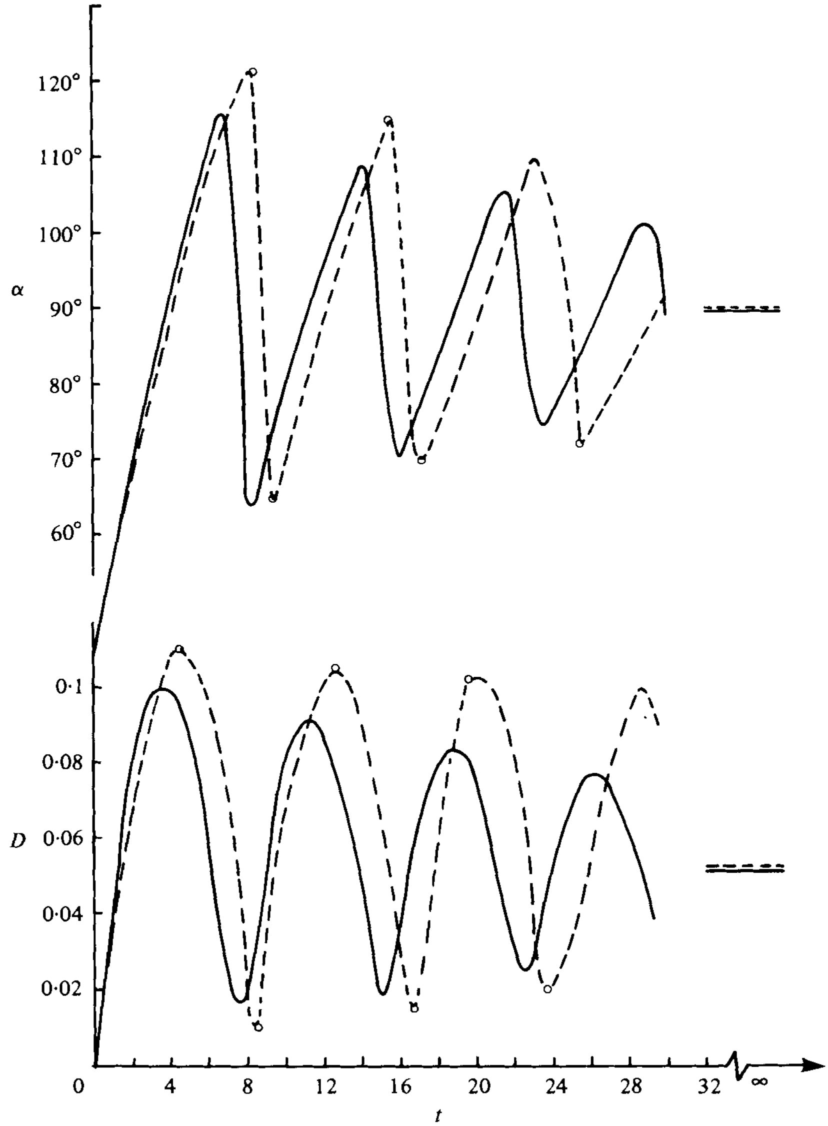

FIGURE 1. The quantities $\alpha(t)$, $D(t)$ for $\lambda = 25$, $k = 0{\cdot}66$. ——, $O(\epsilon)$ theory (8), (9); - - -, experiments of Torza *et al.* (1972).

4. High viscosity with comparably weak flow: $k, \lambda \gg 1$

We consider finally the case when k and λ are comparably large. Thus $k = \beta\epsilon^{-1}$, $\lambda = \epsilon^{-1}$ with $\beta = O(1)$. The $O(1)$ equation can be derived either from the $\lambda \to \infty$, $k \to \infty$ limit, or with the limiting processes reversed, provided, of course, the $O(\epsilon)$ equations are used as an intermediate step. Either route gives

$$\frac{\mathscr{D}\mathbf{F}}{\mathscr{D}t} = \tfrac{5}{6}\mathbf{E} - \tfrac{20}{19}\beta\mathbf{F} + O(\epsilon). \tag{11}$$

This formula plainly cannot be derived from the $O(1)$ equations (7) or (10).

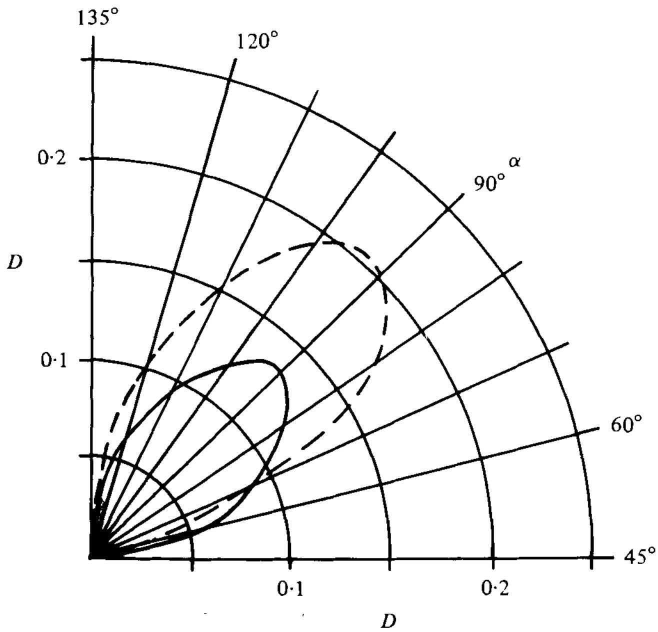

FIGURE 2. Polar plot of D *vs.* α for $\lambda = 21$, $k = 0$. ——, $O(\epsilon)$ theory (8), (9); – – –, experiments of Torza *et al.* (1972). Period of one oscillation: theory, 7·9; experiment, 8·7.

At first sight, consideration of the error terms in, say, (8)′, (9)′ would suggest that (11) is as accurate a result as may be written down in this double limit. In fact, however, the terms of $O(k/\lambda^2)$ in (8)′ are known from (3), and hence (11) may be improved by the $O(\epsilon)$ theory:

$$\frac{\mathscr{D}^*\mathbf{F}}{\mathscr{D}t} = \tfrac{5}{6}\mathbf{F} - \tfrac{20}{19}\beta\mathbf{F} + \epsilon\left[\tfrac{10}{7}Sd(\mathbf{E}.\mathbf{F}) - \tfrac{5}{4}\mathbf{E} + \tfrac{510}{361}\beta\mathbf{F} + \tfrac{9864}{2527}\beta Sd(\mathbf{F}^2)\right] + O(\epsilon^2), \tag{12}$$

$$\frac{\mathscr{D}^*\mathbf{H}}{\mathscr{D}t} = \tfrac{1}{14}Sd_4(\mathbf{EF}) - \tfrac{36}{17}\beta\mathbf{H} - \tfrac{8}{4645}\beta Sd_4(\mathbf{FF}) + O(\epsilon). \tag{13}$$

Equations (8)′, (9)′ may be recovered at once by setting $\beta = 0$, and hence (12), (13) are uniformly valid in k as $\lambda \to \infty$.

Cox (1969) considered the behaviour of the solution trajectories of (11), and showed the existence of an equilibrium within the range of validity of the analysis. In the previous section it was noted that, even when the $O(\epsilon)$ terms for the flow are included, the strength of the flow is underestimated in regard to the distortion it produces. In this limit, however, we find that the inclusion of $O(\epsilon)$ terms for the large surface tension tends also to underestimate the strength of the restoring force, with the net effect that the magnitude of the distortion is significantly increased over that predicted by (11), and can be larger than that found experimentally.

Figure 3 shows a typical time evolution for a case considered experimentally by Torza *et al.* (1972). The maximum deformation (0·3) is only 3 % below that found experimentally, compared with 20 % for the $O(1)$ theory. The period, however, is now overestimated. In figure 4 we show a less successful case with $\lambda = 4{\cdot}3$. Here the $O(1)$ theory shows the correct qualitative behaviour, but considerably underestimates the

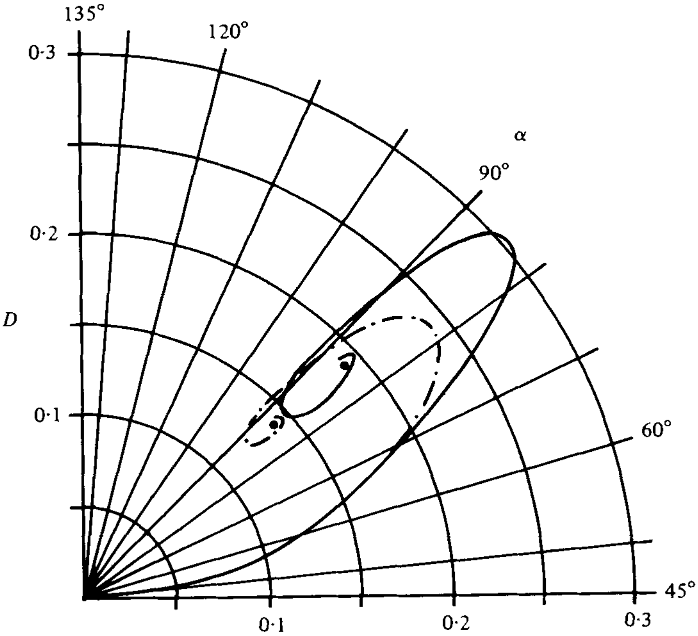

FIGURE 3. Polar plot of D *vs.* α for $\lambda = 8{\cdot}8$, $k = 0{\cdot}81$, $\beta = 0{\cdot}09$. ——, $O(\epsilon)$ theory (12), (13); –·–·–, $O(1)$ theory (11). Maximum value of D attained: $O(\epsilon)$ theory, 0·24; $O(1)$ theory, 0·30; experiment, 0·31. Time elapsed between first and second maximum: $O(\epsilon)$ theory 19·6; $O(1)$ theory, 8·6; experiment, 13·0.

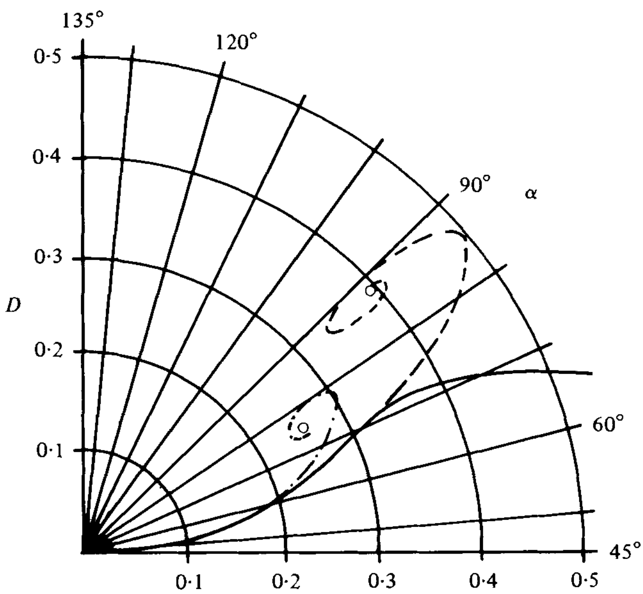

FIGURE 4. Polar plot of D *vs.* α for $\lambda = 4{\cdot}3$, $k = 2{\cdot}1$, $\beta = 0{\cdot}48$. ——, $O(\epsilon)$ theory (12), (13); –·–, $O(1)$ theory (11); – – –, experiments of Torza *et al.* (1972).

deformation. The $O(\epsilon)$ theory follows the $D(t)$ curve from sphericity to a larger deformation (0·35), but then diverges from it, becomes unbounded, and no equilibrium is found. Plainly $\lambda = 4{\cdot}3$ is not sufficiently large for the $\lambda \gg 1$ theory to be appropriate.

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