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The determination of the viscosity coefficients of nematic liquid crystals

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This paper considers the flow generated by driving a sample of nematic liquid crystal through a rectangular capillary by application of a small pressure gradient in the presence of a large aligning magnetic field. A theoretical calculation based on the continuum theory of nematics is presented which makes some allowance for non-uniform alignment induced by flow, and allows a more accurate determination of the viscosities corresponding to the three principal configurations in the plane of shear.

The continuum theory proposed by Ericksen [1] and Leslie [2] describes many of the physical properties of nematic liquid crystals rather well (see for example Chandrasekhar [3], Leslie [4] and de Jeu [5]). The theory contains six viscosity coefficients in the dissipative part of the stress tensor; these are commonly denoted by $\alpha_1, \dots, \alpha_6$. Leslie [4, 6] has derived inequalities upon these coefficients necessary to ensure that entropy production is positive. Parodi [7] has imposed a further restriction upon the α s in the shape of an Onsager relation

$$\alpha_6 - \alpha_5 = \alpha_3 + \alpha_2, \quad (1)$$

which reduces the number of independent coefficients to five, and Currie [8, 9] has presented arguments which support the need for this condition. In a more recent paper, Vertogen [10] has presented an alternative derivation of the nematic theory that contains five independent viscosity coefficients consistent with previous arguments. However, he makes the additional assumption that the stress is derivable from a dissipation function, and that this should essentially produce the Parodi relation is a result anticipated by Ericksen [11]. It is of interest to determine the values of these coefficients for various nematic materials. In addition, applications in electro-optic display devices clearly demonstrate the need for detailed information concerning these viscosity coefficients, this being essential for a complete understanding of the complex coupling between flow and alignment in transient processes in liquid crystal cells.

Most experimental techniques designed to measure the viscosities of nematics employ various means to align the sample in the different directions relative to the flow. Externally applied electromagnetic fields exert body torques which tend to align the director either parallel or perpendicular to the direction of the field. In the absence

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of field effects it is observed that for most nematic liquid crystals the flow itself tends to align the bulk of the sample at a given angle (θ_0) to the streamlines. However, there are materials which do not flow align over their entire nematic temperature range [12–14] and this results in a rich variety of flow induced instabilities (see, for example, Clark *et al.* [15]). Finally by prior treatment of the bounding surfaces it is possible to dictate particular orientations of the director at such boundaries, and these appear relatively insensitive to the presence of other aligning influences. It is usually assumed in such viscometric experiments that an electromagnetic field when present assumes the dominant role, being sufficiently strong to align the sample completely in the required direction. A test of sufficient strength is that the values of the coefficients obtained approach some limit as the magnitude of the field is increased. However in practice this is not always the case, boundary and end effects may be significant. These depend upon the geometry and dimensions of the experimental apparatus in use. In addition, the applied fields, although large, are finite and the lack of related theoretical calculations to guide extrapolation of experimental data makes interpretation of the results uncertain. Consequently the literature contains a rather wide spread of values for the viscosity coefficients.

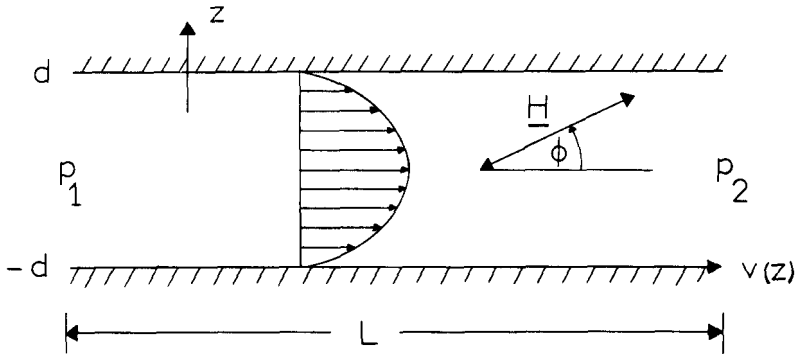


Figure 1. Illustration of the flow considered. A pressure gradient $a = (p_1 - p_2)/L > 0$ (where L is the length of the capillary) drives the sample of nematic. A strong magnetic field at a constant angle ϕ ($\in [0, \pi/2]$) to the flow aligns the anisotropic axis.

Of the classical viscometric flows perhaps the simplest is that of plane Poiseuille flow, where the fluid is driven through a rectangular capillary by an applied pressure gradient and the flux of fluid over a certain time interval is measured (see figure 1). The viscosity $\bar{\eta}$ is then defined in a very natural way by the relation

$$\bar{\eta} = \frac{2ad^3}{3Q}. \quad (2)$$

Here a is the pressure gradient, d half the gap width and Q the constant flux per unit time per unit breadth given by

$$Q = \int_{-d}^d v(z) dz, \quad (3)$$

where $v(z)$ is the velocity in the direction of a and z the coordinate normal to the plates.

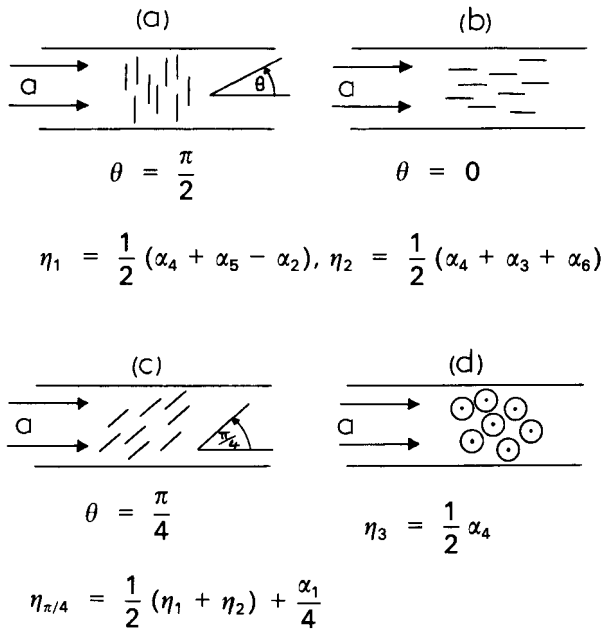


Figure 2. Configurations corresponding to the four principal viscosities: (a), (b), (c) in the plane of the flow and (d) normal to this plane. Associated viscosities are given in the notation of Helfrich [16].

In an identical way a sample of nematic may be subject to such a flow. In this case, when the director is uniformly aligned in the plane of shear at an angle θ to the lower boundary (see figure 2(a)) the viscosity is given by $g(\theta)$, where

$$2g(\theta) = 2\alpha_1 \sin^2 \theta \cos^2 \theta + (\alpha_5 - \alpha_2) \sin^2 \theta + (\alpha_3 + \alpha_6) \cos^2 \theta + \alpha_4. \quad (4)$$

Three measurements in the plane of the flow (see figure 2(a), (b), (c)) can be taken to determine various combinations of the α s. With the director oriented normal to this plane (figure 2(d)) the α_4 coefficient may be evaluated. Note that this latter configuration is unstable above a critical pressure gradient, as discussed by Janossy *et al.* [17]. Henceforth let us restrict attention to the in-plane alignments. Finally it can be shown [4] that provided the material flow aligns, the flow alignment angle θ_0 is related to the α s through

$$\tan^2 \theta_0 = \frac{\alpha_3}{\alpha_2}. \quad (5)$$

This angle may be determined from optical checks on the alignment of the director of the nematic material subject to such a flow and gives the fifth equation from which the viscosity coefficients are readily calculated. As with isotropic fluids, the quantity that is measured experimentally is the flux per unit time and this may be written in terms of the various material parameters of the theory. The apparatus is calibrated with liquids of known viscosity and in this case $\bar{\eta}$ in expression (2) essentially defines the apparent or effective viscosity of the fluid, *and is the value actually measured*. For a uniformly aligned sample of nematic, the viscosity corresponding to a given configuration will be identically equal to $\bar{\eta}$. However for large but finite applied fields

Table 1. Comparison of anisotropic viscosities of MBBA at $\sim 25^\circ\text{C}$.

Investigator (method)	η_1/cP $\theta = \pi/2$	η_2/cP $\theta = 0$	$\alpha_1 \equiv \eta_{12}/\text{cP}$ $\theta = \pi/4$
Gähwiler [18, 19] (capillary flow)	103.5 ± 1.5	23.8 ± 0.3	6.5 ± 4
Summerford <i>et al.</i> [24] (shear stress measurements on a suspended plate)	138 ± 3	—	—
Martinoty and Candau [25] (reflexion of ultrasonic shear waves)	—	~ 27	—
Kneppe and Schneider [22] (capillary flow)	136.1	23.9	-18.1
Kneppe <i>et al.</i> [26] (rotating magnetic field plus results of [22])	135.5	23.9	-18.1

there is an error due to the non-uniformity of the director, and this we endeavour to quantify.

Gähwiler [12, 18, 19] was the first to obtain a complete set of values of the coefficients by this method. More recent attempts using variants of the above include those of Skarp *et al.* [20], Kim *et al.* [21], Kneppe and Schneider [22] and Beens and de Jeu [23].

For the nematic 4-methoxybenzylidene-4'-*n*-butylaniline (MBBA) the results of several investigations by a variety of techniques are given in table 1. Using the attenuation of ultrasound, Martinoty and Candau [25] actually measured the quantity

$$\tilde{\eta}_2 = \eta_2 - \frac{\alpha_2 \alpha_3}{\gamma_1} \tan^2 \theta_0, \quad (6)$$

where $\gamma_1 = \alpha_3 - \alpha_2$. Since the flow alignment angle for MBBA at 25°C is approximately 6° [12, 27] it follows that $\tilde{\eta}_2$ is a good estimator of η_2 in this case. The results of Kneppe *et al.* [26] were obtained by application of a rotating magnetic field to determine γ_1 . Using the values of η_2 from [22] and θ_0 from [27] they calculate η_1 through the relation

$$(\eta_1 - \eta_2) \cos 2\theta_0 = \gamma_1, \quad (7)$$

thus avoiding the uncertainty of a direct measurement of the η_1 coefficient. As is evident from table 1, there is good agreement in the results for η_2 . However for η_1 (see figure 2(a)) and α_1 there is a marked disparity. The latter viscosity is determined from the algebraic equation

$$\alpha_1 = 4\eta_{\pi/4} - 2(\eta_1 + \eta_2), \quad (8)$$

and so the accuracy of the result is highly dependent on the estimate of η_1 . The variation in the η_1 values quoted in the table is essentially the reason for the disagreement in the α_1 coefficient. Doubt as to the validity of Gähwiler's results for η_1 were raised initially by Summerford *et al.* [24] and discussed more fully by de Jeu [28]. They argue that for the pressure gradients used by Gähwiler ($40\text{--}200 \text{ dyn cm}^{-3}$), the

applied magnetic fields (up to 6 kG) are insufficient to overcome the strong flow aligning effects and orient the anisotropic axis parallel to the direction of the field. Since the flow alignment angle for this temperature is of the order of 6°, when measuring η_2 (see figure 2(b)), viscous and field effects tend to compete weakly. However as θ increases, so the field strengths, required to dominate the large viscous torques present, must also increase. The smaller value of η_1 obtained by Gähwiler would tend to support the idea that even for a magnetic field of 6 kG, the director in this experiment is aligned at an angle somewhat less than $\pi/2$ radians to the flow. Here we present a theoretical calculation to resolve this discrepancy.

Consider the arrangement illustrated in figure 1. The sample of nematic is contained between two parallel semi-infinite plates of gap width $2d$. A pressure gradient a drives the flow in one direction, and a magnetic field \mathbf{H} is applied in the plane of shear at a constant angle $\phi (\in [0, \pi/2])$ to the lower boundary. As Raynes *et al.* [29] describe, it is feasible to arrange the inclination of the optic axis on a solid surface at any prescribed angle. Thus by setting boundary conditions on the director compatible with the orientation of the field, it is possible to isolate the competition between the flow and the magnetic field to align the director. In practice, the effect of a strong anchoring boundary condition is confined to within two thin transition regions near the solid surfaces. Gähwiler [19] finds that the thickness of these layers for his experiments is between 3 per cent and 6 per cent (for η_1 with an applied field of 9 kG) of the plate separation. However, Beens and de Jeu [23] demonstrate that this boundary layer effect is rather important in the measurement of η_1 . The appropriate form of the continuum equations describing the spacial variation of the director and the velocity (assuming steady flow) is written as

$$2f(\theta)\theta'' + \frac{df}{d\theta}\theta'^2 + \frac{2am(\theta)z}{g(\theta)} + \chi_a H^2 \sin 2(\phi - \theta) = 0, \tag{9 a}$$

$$v'(z) = \frac{-az}{g(\theta)}, \tag{9 b}$$

where

$$f(\theta) = k_1 \cos^2 \theta + k_3 \sin^2 \theta, \tag{10 a}$$

$$m(\theta) = \alpha_3 \cos^2 \theta - \alpha_2 \sin^2 \theta \tag{10 b}$$

and $g(\theta)$ is given by expression (4). χ_a in equation (9 a) represents the diamagnetic anisotropy, which is taken to be constant and positive, while k_1 and k_3 in equation (10 a) are the splay and bend elastic constants respectively. All primes denote differentiation with respect to z . For boundary conditions take

$$\left. \begin{aligned} \theta(-d) &= \theta(d) = \phi, \\ v(-d) &= v(d) = 0, \end{aligned} \right\} \tag{11}$$

the latter being the familiar no-slip assumption on the velocity. One basic solution to equations (9) and (11) for all values of ϕ , is the uniform equilibrium state

$$\theta \equiv \phi, \quad v \equiv 0 \quad (a = 0), \tag{12}$$

in which there is no flow, and the director is aligned parallel to the applied field. By solving the corresponding linearized version of equations (9) it may be shown that for sufficiently small a and large H ($\equiv |\mathbf{H}|$, the magnitude of the field), an appropriate

form of solution is given by

$$\theta(z) \sim \phi + \psi(z), \quad (13 a)$$

where

$$\psi(z) = \delta \left[\frac{z}{d} - \frac{\sinh(\xi z/d)}{\sinh \xi} \right] \quad (13 b)$$

and the velocity assumes a parabolic profile

$$v(z) \sim \frac{a}{2g(\phi)} [d^2 - z^2]. \quad (14)$$

Here the dimensionless quantities ξ and δ are of the form

$$\xi^2 = \frac{d^2 \chi_a H^2}{f(\phi)} \gg 1, \quad (15 a)$$

$$\delta = \frac{m(\phi) ad}{g(\phi) \chi_a H^2} \ll 1. \quad (15 b)$$

Integration of expression (3) by parts once, and use of boundary conditions (11) yields

$$Q = - \int_{-d}^d z v'(z) dz. \quad (16)$$

If one substitutes equation (9 b) into equation (16), the flux may be written as

$$Q = 2a \int_0^d \frac{z^2}{g(\theta)} dz. \quad (17)$$

We expand $g(\theta)$ in a three term Taylor series about the uniform state $\theta = \phi$

$$g(\theta) \sim g(\phi) + \psi \left. \frac{dg}{d\theta} \right|_{\theta=\phi} + \frac{\psi^2}{2} \left. \frac{d^2g}{d\theta^2} \right|_{\theta=\phi} \quad (18)$$

and since $dg/d\theta|_{\theta=\phi}$ is zero for $\phi = 0, \pi/2$, approximate (18) by the two term truncated series

$$g(\theta) \sim g(\phi) + \frac{\psi^2}{2} \left. \frac{d^2g}{d\theta^2} \right|_{\theta=\phi}, \quad \phi = 0, \pi/2, \quad (19 a)$$

$$g(\theta) \sim g(\phi) + \psi \left. \frac{dg}{d\theta} \right|_{\theta=\phi}, \quad \phi \neq 0, \pi/2. \quad (19 b)$$

If expressions (19) are substituted into the integrand of equation (17), together with the form of $\psi(z)$ given in equation (13 b), evaluation of the integral neglecting terms whose magnitude is $O(\xi^{-1})$ or less in the limit as $\xi \rightarrow \infty$, leads to

$$\left. \begin{aligned} Q &\sim \frac{2ad^3}{3g(\phi)} \left[1 - \frac{6\delta^2[\alpha_1 \cos 4\phi - \gamma_2 \cos 2\phi]}{5g(\phi)} \right], & \phi = 0, \pi/2, \\ Q &\sim \frac{2ad^3}{3g(\phi)} \left[1 - \frac{3\delta[\alpha_1 \sin 4\phi - 2\gamma_2 \sin 2\phi]}{8g(\phi)} \right], & \phi \neq 0, \pi/2, \end{aligned} \right\} \quad (20)$$

where $\gamma_2 = \alpha_6 - \alpha_5$ ($\equiv \alpha_3 + \alpha_2$ by equation (1)). Finally, combining equations (2) and (20) it follows that

$$\frac{g(\phi)}{\bar{\eta}} \sim 1 - \begin{cases} \frac{6\delta^2[\alpha_1 \cos 4\phi - \gamma_2 \cos 2\phi]}{5g(\phi)}, & \phi = 0, \pi/2, \\ \frac{3\delta[\alpha_1 \sin 4\phi - 2\gamma_2 \sin 2\phi]}{8g(\phi)}, & \phi \neq 0, \pi/2. \end{cases} \quad (21)$$

Thus to our approximation this yields a quadratic equation in $g(\phi)$

$$\left. \begin{aligned} 5g^2(\phi) - 5g(\phi)\bar{\eta} + 6\delta^2[\alpha_1 \cos 4\phi - \gamma_2 \cos 2\phi]\bar{\eta} &= 0, & \phi = 0, \pi/2, \\ 8g^2(\phi) - 8g(\phi)\bar{\eta} + 3\delta[\alpha_1 \sin 4\phi - 2\gamma_2 \sin 2\phi]\bar{\eta} &= 0, & \phi \neq 0, \pi/2, \end{aligned} \right\} \quad (22)$$

and the appropriate root is given by

$$g(\phi) = \frac{5\bar{\eta} + \sqrt{\{25\bar{\eta}^2 - 120\delta^2[\alpha_1 \cos 4\phi - \gamma_2 \cos 2\phi]\bar{\eta}\}}}{10}, \quad \phi = 0, \pi/2, \quad (23 a)$$

$$g(\phi) = \frac{8\bar{\eta} + \sqrt{\{64\bar{\eta}^2 - 96\delta[\alpha_1 \sin 4\phi - 2\gamma_2 \sin 2\phi]\bar{\eta}\}}}{16}, \quad \phi \neq 0, \pi/2. \quad (23 b)$$

It is worth noting at this stage that for a given pressure gradient, as $H \rightarrow \infty$, then $\xi \rightarrow \infty$ and $\delta \rightarrow 0$. From equations (13) and (21) or (23) we conclude that in this limit

$$\left. \begin{aligned} \theta(z) &\rightarrow \phi, \\ g(\phi) &\rightarrow \bar{\eta}. \end{aligned} \right\} \quad (24)$$

representing a uniformly aligned sample parallel to the field \mathbf{H} . In addition, by setting $\phi \equiv \theta_0$ (when this angle exists), it is clear from equations (5) and (10 b) that $m(\phi) \equiv 0$ and so from expression (15 b), $\delta \equiv 0$, again reproducing the result in equation (24). This is not surprising since in this case flow, field and surface influences *all* tend to align the anisotropic axis in the same direction.

Expressions (21) enable the calculation of the extrapolation factor in measuring any of the in-plane viscosities to be made, and using this, provided it is sufficiently small, to approximate the limiting value of the viscosity. However to compute these extrapolation factors, estimates of the α s are required. We suggest the following approach. For a strong magnetic field and small pressure gradient, the viscosity function $g(\phi)$ will be constant *to leading order* and given in terms of the flux Q by expression (2). The four measurements corresponding to figures 2(a)–(d) are taken, resulting in four algebraic equations for the α s. Combine these with the Parodi relation (1) and a measurement of the flow alignment angle (cf. equation (5)) in the absence of the field. Solving these six equations gives *to the same order of magnitude*, estimates for $\alpha_1, \dots, \alpha_6$ and these may be used in the evaluation of the extrapolation factors. The latter are then applied to refine the original estimates via equations (21). From physical reasoning it is expected that for a given field strength and pressure gradient, the largest error associated with the non-uniform alignment of the director to correspond to the case of $\phi = \pi/2$ (η_1 configuration). However from equations (21), this is seen not to be the case, since, for $\delta \ll 1$, $\delta^2 \ll \delta$ and so intermediate values of ϕ produce the greatest error. This results from the original series truncation in (19) since the first derivative of $g(\theta)$ vanishes at $\theta = 0, \pi/2$. Therefore, to extrapolate

Table 2. For a typical pressure gradient of 140 dyn cm^{-3} , and an aligning field of 6 kG , the errors (from expressions (21 a) and (21 b)) associated with measuring the three principal in-plane viscosities are listed. A negative sign indicates that the measured value of the viscosity is lower than the actual value (calculations based on the data and experimental details supplied in [12, 18, 19]).

Angle of field	$\phi = 0$ (η_2)	$\phi = \frac{\pi}{4}$ ($\eta_{\pi/4}$)	$\phi = \frac{\pi}{2}$ (η_1)
$\frac{6\delta^2[\alpha_1 \cos 4\phi - \gamma_2 \cos 2\phi]}{5g(\phi)}$	0.006	—	-0.284
$\frac{3\delta[\alpha_1 \sin 4\phi - 2\gamma_2 \sin 2\phi]}{8g(\phi)}$	—	0.433	—

in a meaningful way for $\phi \in (0, \pi/2)$, a , d and H must be such that

$$\left| \frac{\delta(dg/d\theta)}{g(\theta)} \right|_{\theta=\phi} \ll 1. \tag{25}$$

Table 2 lists the errors associated with measuring the three principal in-plane viscosities for a typical pressure gradient of 140 dyn cm^{-3} and an aligning field of 6 kG (the maximum applied in the Gähwiller experiments). Clearly for η_2 the error is less than 1 per cent and consequently the value quoted for this coefficient is in excellent agreement with the results of the other investigations. However this is not the case with η_1 and $\eta_{\pi/4}$. For the latter viscosity, the magnitude of the error is too large to satisfy condition (25). When $\phi = \pi/2$, $\bar{\eta} = 103.5$ [18] and a more accurate estimate of η_1 may be obtained from equation (23 a)

$$\begin{aligned} \eta_1 &= \frac{5\bar{\eta} + \sqrt{\{25\bar{\eta}^2 - 120\delta^2[\alpha_1 + \gamma_2]\bar{\eta}\}}}{10}, \\ &= 127.4 \text{ cP}. \end{aligned} \tag{26}$$

This calculation was based on the values of viscosity coefficients supplied in [18]. The result is more in accord with the outcome of the Knepe and Schneider experiment. Their value of 136.1 cP is not reproduced exactly in this calculation for essentially two reasons. Firstly, the flow rates used by Gähwiller are up to six times larger than those of Knepe and Schneider and for these larger flow rates, the assumptions (a small, H large) on which our analysis is based are only approximately satisfied. Hence the extrapolation factor of -0.284 (table 2) is large in magnitude. Secondly, there is an error associated with truncating the Taylor series for $g(\theta)$ in equation (19 a), although this latter effect is less marked.

Similarly the extrapolation factors may be calculated from equation (21) for the experiment of Knepe and Schneider. For a typical pressure gradient of 50 dyn cm^{-3} , the extrapolation factor in measuring η_1 (with a 6 kG field) is of the order of 9 per cent, consistent with their claim. They present a related calculation (not based on the continuum theory) to enable them to approximate the viscosities in the limit of infinite magnetic induction.

Finally, for a given calibration error of 10 per cent, table 3 lists the minimum field and maximum pressure gradient allowable to sustain the alignment of the director in

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Table 3. Minimum field ($a = 140 \text{ dyn cm}^{-3}$) and maximum pressure gradient ($H = 6 \text{ kG}$) for a given calibration error of 10 percent. Calculations based on details supplied in [19].

Angle of field	$\phi = 0$ (η_2)	$\phi = \frac{\pi}{4}$ ($\eta_{\pi/4}$)	$\phi = \frac{\pi}{2}$ (η_1)
Minimum field/kG $a = 140 \text{ dyn cm}^{-3}$	2.950	12.480	7.790
Maximum pressure gradient (dyn cm^{-3}) $H = 6 \text{ kG}$	580	33	83

the three configurations within the plane of the flow. These are calculated once again from expressions (21). Under the experimental conditions described by Gähwiler, η_2 is determined to within the required accuracy, while η_1 and $\eta_{\pi/4}$ are clearly not.

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