# Simultaneous Measurement of Viscosity and Density with an Oscillating-Disk Instrument: The Effect of Fixed Plates

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The period and damping of the free motion of a body oscillating in a fluid depend on the fluid's viscosity and density. Commonly, a working equation which expresses the damping as a function of the viscosity and density is solved for the viscosity, the damping being measured and the density being treated as an independently supplied parameter. Another working equation exists for the period, and, in general, the period depends on a combination of the viscosity and the density which is linearly independent of the combination that appears in the damping equation. It is, therefore, in principle, possible to determine both the viscosity and the density by a simultaneous solution of the two coupled working equations, since the period also is measured. In this paper, the working equations that describe the oscillating-disk viscometer are reviewed and their simultaneous solution is considered. The effect of fixed plates symmetrically located above and below the oscillating disk is of specific interest. The paper's main result is that fixed plates can dramatically increase the independence of the damping and period working equations, so that it becomes indeed feasible to determine the viscosity and the density of a fluid simultaneously from the damping and period of oscillating motion. A price is paid, however, because the instrument's working equations when plates are present have multiple solutions. Under special conditions, these multiple solutions can coalesce, and then one can only deduce the viscosity from the damping equation if the density is known a priori.

**KEY WORDS:** densimeter; oscillating-disk viscometer; simultaneous viscosity and density measurement; toluene; viscometer; viscosity.

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# 1. INTRODUCTION

Some of the most commonly used methods to measure the viscosity of a fluid require that the fluid's density be known. This is true, for example, for the gravity-driven capillary-flow method [1] and the falling-body method [2]. In these methods, one measures directly a single quantity (time of efflux of a certain volume of fluid in the capillary-flow method, and a body's fall time over a certain distance in the case of the falling-body method) which is related to the viscosity and density by a working equation. The traditional approach has been to include in the viscometer a provision for measuring the fluid's temperature and pressure. The density can then be calculated from an equation of state, if one is available. Alternatively, the experimenter can make a parallel measurement of the density using a densimeter charged with another sample of the fluid under study and operated at the same temperature and pressure. Currently there is interest in simultaneous measurements of the viscosity and the density. By this we mean measurements of the two properties made in the same instrument, on the same sample, at the same time. With simultaneous methods, the experimenter is free to study fluids for which an accurate equation of state is not available. Simultaneous measurements have an advantage also over parallel measurements of the two properties, since they eliminate errors involved with producing identical conditions of temperature and pressure in separate instruments.

One way to achieve simultaneous measurements is to build a device that incorporates conceptually distinct density and viscosity measurement methods into one apparatus. For example, Wagner and coworkers [3, 4] used a nearly frictionless magnetic coupling to levitate a cylindrical sinker which is immersed in the fluid under study. The force necessary to levitate the sinker is less than its known weight by the buoyant force exerted on it by the fluid. Since the sinker volume is known, the fluid density can be determined. Furthermore, a rotary motion can be imparted to the sinker, the magnetic suspension acting then as a bearing. When an external driving torque is switched off, the rotary motion decays exponentially. The decay rate is a function of the fluid's viscosity and density, so the viscosity can be determined. Greer and coworkers [5, 6] used a similar device, but they imparted a steady motion to the sample cell while holding the sinker at a fixed position magnetically. Veliyulin and coworkers [7] used an oscillating-disk viscometer in which the suspension wire hung from an electronic balance, enabling measurement of the buoyant force.

We note that in these methods, the density and viscosity working equations are not coupled. That is, the viscosity does not appear in the working equation of the density measurement. Indeed, these instruments do not require fluid flow for the measurement of the density.

Another type of simultaneous measurement is possible with viscometers that employ a vibrating body. Examples of vibrating-body viscometers include vibrating-wire viscometers and torsionally vibrating piezoelectric crystal viscometers [8], as well as oscillating-body viscometers [9]. In the case of oscillating-body viscometers, a body is set into an oscillatory motion which is then allowed to decay freely. The motion is characterized by the period and the decay rate of the exponentially damped harmonic motion. On the other hand, in the vibrating-crystal method the crystal is driven at constant amplitude with the aid of an external periodic force of variable frequency. Again, there are two parameters that describe the motion: the resonant period and the width of the resonance curve. Generally, there are two parameters that describe vibratory motion, and they are both affected by the hydrodynamic drag exerted on the body by the fluid. In cases of interest, both motional parameters depend on both fluid properties, density and viscosity. Since both parameters can be measured, vibrating-body viscometers can, in principle, provide simultaneous measurements of the density and viscosity. The working equations are coupled, however. That is, two equations each relate the density and viscosity with the two motional parameters, and these equations are typically rather complicated.

Padua and coworkers [10–12] have made simultaneous density and viscosity measurements with vibrating-wire viscometers. The coupled working equations describing such instruments are formally quite similar to those pertaining to the oscillating-disk viscometer, which we will provide in the next section. Padua and coworkers, however, employed an ingenious design that decreases the coupling and greatly increases the precision of their instrument's density measurements. That is, they arranged for the tension in the wire to be provided by the weight of a sinker immersed in the fluid. This design causes the wire's vibration period to have an extra fluid density dependence (due to the buoyancy of the sinker, independent of the viscosity) in addition to the density dependence introduced through hydrodynamic drag (which depends on both fluid properties). A precision for the density at the level of 0.05% was attained, comparable to that provided by stand-alone densimeters, while the viscosity was measured with a precision of 0.5% [11, 12].

We have used an oscillating-disk viscometer to make simultaneous measurements of viscosity and density [13]. In an oscillating-disk instrument, the dependence of the motional parameters on the fluid properties is due to hydrodynamic drag only. A consequence is that the density and viscosity are measured with approximately equal precision. In Ref. 13, we attained a precision of about 0.2% for the simultaneously measured viscosity and density of liquid toluene over a wide range of temperatures and pressures.

Krall and Sengers

The period and damping measurements upon which Ref. 13 was based also provided the basis of a set of experimental viscosities for liquid toluene at pressures up to 30 MPa [14]. However, in Ref. 14, we did not measure the density simultaneously. Instead, we calculated the density of toluene from an equation of state [15], and determined the viscosity of toluene from the measured damping. Thus in Ref. 14 we worked only with one of the instrument's working equations, namely the one sensitive to the damping. As we showed in Ref. 13, the agreement of the viscosity found in this way with the viscosity as measured simultaneously with the density was at the level of 0.4%. Agreement of the simultaneously determined density with the density predicted by the equation of state was comparable [13].

The emphasis of Ref. 13 was primarily on experimental details. In this paper, we discuss some mathematical aspects of simultaneous measurements with oscillating-disk viscometers, in particular, the effects of the often-used fixed plates, which have not been noticed before in the literature [13, 16, 17]. We also include here an experimental section summarizing the measurements of Ref. 13 and comparing them to more recent recommended interpolating equations for the properties of toluene.

# 2. WORKING EQUATIONS FOR AN OSCILLATING-DISK VISCOMETER

In an oscillating-disk viscometer, a disk hangs from a suspension wire. The disk oscillates about its axis, the wire supplying a linear restoring torque. When in vacuo, the disk's angular displacement from the rest position,  $\alpha(t)$ , is given by [17]

$$\alpha(t) = A_0 \exp(-\Delta_0 \omega_0 t) \sin(\omega_0 t) \tag{1}$$

and, when in a fluid, by

$$\alpha(t) = A \exp(-\Delta \omega t) \sin(\omega t).$$
<sup>(2)</sup>

Here  $\Delta$  is the damping in the fluid and  $\Delta_0$  the damping in vacuo. The oscillation periods are  $T = 2\pi/\omega$  in the fluid and  $T_0 = 2\pi/\omega_0$  in vacuo. The amplitudes A and  $A_0$  do not enter into the working equations, and also need not be measured since techniques exist for determining the period and damping from time-interval measurements.

As is discussed in, for example, Refs. 9 and 17,  $\Delta$  and T are related to the fluid's density  $\rho$  and viscosity  $\eta$  through

$$(s + \Delta_0)^2 + 1 + D(s) = 0, (3)$$

where  $s = (-\Delta + i) \Theta$  with  $\Theta = T_0/T$  and  $i = \sqrt{-1}$ ; D(s) is the Laplace transform of the viscous torque exerted on the disk by the fluid. To obtain an explicit expression for D(s), one needs to solve a partial differential equation subject to boundary conditions on all surfaces. There is no known exact solution for the two-dimensional problem corresponding to a disk (axial symmetry eliminates one dimension). A first approximation to D(s)is found by considering infinite oscillating bodies which yield an easy-tosolve one-dimensional problem. Thus, one estimates the torque on each of the two flat surfaces of the disk by calculating the torque exerted on a circular region, of radius R and co-axial with the axis of oscillation, contained within an oscillating plane of infinite radial extent. Likewise, the torque on the cylindrical surface is estimated as the torque on a cylindrical section, of height 2h, contained on a cylinder of radius R and of infinite axial extent. The simple addition of these two estimates gives an approximation to D(s)which neglects the "edge effect" due to the deviation of the fluid flow pattern in the vicinity of the rims of the disk from the flow patterns near either of the infinite surfaces. For a more accurate approximation, one may then evaluate the edge effect as a perturbation, as has been done by Azpeitia and Newell [16]. For D(s) we use the approximation,

$$D(s) = \frac{\rho\delta}{\bar{\rho}h} \left\{ \left[ \coth\left(\frac{\beta}{\delta}s^{1/2}\right) + 4\frac{h}{R} \right] s^{3/2} + B\left(\frac{\delta}{R}\right)s + C\left(\frac{\delta}{R}\right)^2 s^{1/2} \right\}, \quad (4)$$

where

$$B = \frac{16}{3\pi} \left( \frac{4\pi}{\sqrt{27}} - 1 \right) + 6 \frac{h}{R} \quad \text{and} \quad C = \frac{17}{9} + \frac{3}{2} \frac{h}{R}.$$
 (5)

This expression describes an instrument with fixed plates a distance  $\beta$  above and below the disk. The disk is made of a material that has a density  $\bar{\rho}$ . The boundary-layer thickness  $\delta = (\eta/(\rho\omega_0))^{1/2}$  characterizes the distance from the disk over which there is appreciable fluid flow. The hyperbolic cotangent function pertains to the one-dimensional problem of an infinite plane oscillating below an infinite fixed plane. The terms with coefficients 4h/R, 6h/R, and 3h/(2R) arise from an expansion of the solution, exactly expressible in terms of Bessel functions, which pertains to the one-dimensional problem of an infinitely long cylinder. The terms with coefficients  $\frac{16}{3\pi}(\frac{4\pi}{\sqrt{27}}-1) = 2.408$  and 17/9 pertain to the edge effect of a free disk [16], i.e., one with the fixed plates either absent or removed to a great distance  $\beta \gg \delta$  such that the hyperbolic cotangent function attains the limit of unity. For a free disk, therefore, Eq. (4) gives D(s) correctly to

second order in powers of  $(\delta/R)^2$  and so furnishes the basis for absolute measurements when  $R \gg \delta$  [18].

In Eq. (4), the hyperbolic cotangent function describes a disk between fixed plates, but Eq. (5) implements the edge effect of a free disk. We accept this inconsistency because no expression is available for the edge effect for the case where fixed plates are present with  $\beta$  comparable to  $\delta$ , so that the plates are close enough to the disk to modify the flow, but not so close to allow accurate approximations based on  $\beta \ll \delta$ . (The case of fixed plates with spacing  $\beta \ll \delta$  has been treated by Newell [19].) We find, however, that Eq. (4) with Eq. (5) yields working equations that are accurate to a few percent when applied to our viscometer, even though this instrument has fixed plates at distances of about  $3\delta$  from the disk which do influence the disk's motion and therefore alter the edge effect. In order to achieve better accuracy, we use a calibration procedure in which we adjust the above-mentioned coefficients slightly as guided by measurements we perform in a liquid of known properties. This calibration, which we describe briefly in the experimental section, does not affect any of the qualitative aspects of simultaneous measurements with which we are most concerned in this paper.

The complex equation Eq. (3) with Eq. (4) is most useful when its real and imaginary parts are exhibited explicitly. To do this we follow Ref. 20 and introduce a notation for the real and imaginary parts of the required powers and functions of the complex constant  $s = (-\Delta + i) \Theta$ :

$$s^{1/2} \equiv x + iy \tag{6a}$$

with

$$x = \left\{ \frac{\Theta}{2} \left[ (\Delta^2 + 1)^2 - \Delta \right] \right\}^{1/2} \text{ and } y = \frac{\Theta}{2x}, \tag{6b}$$
$$s^{3/2} \equiv -H_2 + iH_1 \tag{6c}$$

with

$$H_1 = 3\Theta x/2 - \left(\frac{\Theta}{2x}\right)^3$$
 and  $H_2 = 3\Theta^2/4x - x^3$ , (6d)

$$\operatorname{coth}\left(\frac{\beta}{\delta}s^{1/2}\right) \equiv K_2(\delta, \Delta, \Theta) - iK_1(\delta, \Delta, \Theta)$$
 (6e)

with

$$K_1(\delta, \Delta, \Theta) = \frac{\sin(\frac{\theta}{x}\beta/\delta)}{\cosh(2x\beta/\delta) - \cos(\frac{\theta}{x}\beta/\delta)}$$
(6f)

and

$$K_2(\delta, \Delta, \Theta) = \frac{\sinh(2x\beta/\delta)}{\cosh(2x\beta/\delta) - \cos(\frac{\Theta}{x}\beta/\delta)}.$$
 (6g)

For later, it will be helpful to keep in mind that whereas x, y,  $H_1$ , and  $H_2$  are functions of  $\Delta$  and  $\Theta = T_0/T$  only and, therefore, are known quantities once a measurement has been made,  $K_1$  and  $K_2$  are (through  $\delta$ ) functions also of  $\eta$  and  $\rho$  and are, therefore, quantities not known *a priori*. The real and imaginary parts of Eqs. (3) and (4) can now be written explicitly:

$$\Theta^{2} - 1 - (\varDelta \Theta - \varDelta_{0})^{2}$$

$$= \frac{\rho \delta}{\bar{\rho} h} \left\{ [H_{1}K_{1} - H_{2}K_{2}] - 4\frac{h}{R}H_{2} - B\Theta\varDelta\left(\frac{\delta}{R}\right) + Cx\left(\frac{\delta}{R}\right)^{2} \right\}, \quad (7a)$$

$$2(\varDelta \Theta - \varDelta_{0})\Theta$$

$$= \frac{\rho \delta}{\bar{\rho} h} \left\{ [H_{1}K_{2} + H_{2}K_{1}] + 4\frac{h}{R}H_{1} + B\Theta\left(\frac{\delta}{R}\right) + C\frac{\Theta}{2x}\left(\frac{\delta}{R}\right)^{2} \right\}. \quad (7b)$$

It is useful to note that the left-hand side of Eq. (7a) is approximately equal to the negative of twice the fractional "period shift"  $-2(T-T_0)/T$ , while the left-hand side of Eq. (7b) is approximately equal to twice the damping  $2\Delta$ . We therefore refer to Eq. (7a) as the "period" equation and to Eq. (7b) as the "damping" equation. These approximations follow from the facts that  $\Theta$  is close to unity and that  $\Delta_0 \ll \Delta \ll 1$ . In the remainder of this paper we set  $\Delta_0$ , the damping in vacuo, to zero for simplicity.

#### 3. NATURE OF SOLUTIONS OF THE WORKING EQUATIONS

# 3.1. Solution of the Individual Equations for the Viscosity with Specified Density

The quantities  $\Delta$  and  $\Theta = T_0/T$  are measured in an experiment. If the density  $\rho$  is known, either of Eqs. (7) may be solved for  $\delta$ . Then the viscosity may be found from  $\eta = 2\pi\rho\delta^2/T_0$ . We denote by  $\eta_{\text{per}}$  and  $\eta_{\text{damp}}$  the viscosities so determined by, respectively, the period and damping equations.

Now we consider the sensitivity of  $\eta_{per}$  and  $\eta_{damp}$  to small fluctuations dT,  $dT_0$ ,  $d\Delta$  in the values of the periods in fluid and in vacuo, and in the damping:

$$\frac{d\eta_{\rm per}}{\eta_{\rm per}} \simeq \frac{2}{T - T_0} \left( dT - dT_0 \right),\tag{8a}$$

$$\frac{d\eta_{\rm damp}}{\eta_{\rm damp}} \simeq 2 \frac{d\varDelta}{\varDelta} - \frac{dT}{T}.$$
(8b)

These equations show that  $\eta_{damp}$  is negligibly sensitive to  $dT_0$  and sensitive to dT only through the ratio of dT to the total period T. On the other hand,  $\eta_{per}$  is negligibly sensitive to  $\Delta$  but sensitive to both dT and  $dT_0$ according to their ratios to the much smaller quantity  $T-T_0$ . Thus the finite precision with which T can be measured causes much greater imprecision in  $\eta_{per}$  than in  $\eta_{damp}$ . Since modern experimental methods allow time intervals to be measured very accurately and precisely, the fluctuations dTcan be kept so small that the resulting scatter in  $\eta_{per}$  need not present a serious problem in itself. Much more troublesome is the fact that Eq. (8a) shows that the period equation requires the value of  $T_0$  to be known just as accurately as the value of T. This may be difficult because  $T_0$  is not accessible to direct measurement when the instrument is being used to study a fluid. Thus,  $T_0$  must be known from a measurement made at another time, and this value must still be accurate despite, for example, the thermal and pressure cycling to which the instrument has been subsequently subjected. For this reason, when the density  $\rho$  is independently known, the viscosity  $\eta$ is invariably calculated from the damping equation and the period equation is used at most as a consistency check.

With a good instrumental design, however, the period in vacuo,  $T_0$ , can be quite stable. For example, our viscometer, used to collect the data described below, has a vacuum period of about 16.8 s reproducible to within a few ms before and after a typical measurement program, suggesting that the period equation could prove useful. This observation opens the possibility that the damping and the period equations may be solved simultaneously when both the viscosity  $\eta$  and the density  $\rho$  are treated as unknowns. Our main interest in this paper is to discuss some of the qualitative aspects of solving Eqs. (7) for  $\rho$  and  $\eta$  simultaneously which are very different from solving either of them separately for  $\eta$  alone with  $\rho$  specified independently. Clearly, for such a simultaneous method to work it is necessary that Eqs. (7) must in some sense be independent. We shall see that their independence can be quite different depending on whether or not fixed plates are present. Nieuwoudt et al. [17] have considered the situation for a free disk. We begin by reviewing their results, adding some new physical interpretation. We shall then turn to a disk between fixed plates.

#### 3.2. Simultaneous Solutions of Working Equations for a Free Disk

Nieuwoudt et al. [17] have shown that for the free disk in the case that  $R \gg \delta$  holds so strongly that the terms in Eqs. (7) with coefficients *B* and *C* can be neglected, the simultaneous method fails completely. We can see this by inspecting Eqs. (7) (after setting  $K_1 = 0$  and  $K_2 = 1$  as is the case for a free disk) and noting that, with B = C = 0, the fluid properties affect the disk's motion only through the value of the combination  $\rho\delta$ . As a result, in this case  $\Delta$  and *T* are not independent at all: one can be calculated from the other through a relation in which the properties of the fluid do not appear. This relation is

$$1 - \Theta^2 + \varDelta^2 \Theta^2 = 2 \frac{H_2}{H_1} \varDelta \Theta^2, \tag{9}$$

where  $H_1$  and  $H_2$ , both approximately equal to  $1/\sqrt{2}$  as defined in Eqs. (6), depend only on  $\Delta$  and  $\Theta$ . This is the relation that results when  $\rho\delta$ is eliminated from Eqs. (7) in the case of a free disk with B and C set to zero. A useful approximate form of Eq. (9) is the simple expression  $\Theta =$  $1 - \Delta + O(\Delta^2)$  showing that, for a free disk with  $R \gg \delta$  holding so strongly that the higher-order terms in Eqs. (7) (i.e., those with the coefficients B and C) can be neglected, the fractional period shift  $(T-T_0)/T$  approximately equals the damping and, in fact, in this limit can be calculated exactly from the damping (via Eq. (9)) without any information about the fluid having to be specified. Clearly in this situation a measurement of Tcannot provide any more information about the fluid than is present in the value of  $\Delta$ , so the simultaneous method cannot work. Thus, it is seen that, in the case of a free disk, it is required that  $\delta/R$  be large enough that at least the terms with coefficient B make a significant contribution to the right-hand sides of Eqs. (7). For the free disk, Nieuwoudt et al. [17] introduced a small parameter  $b = B(\rho R/(\bar{\rho}h))(\delta/R)^2$  which characterizes the "free" part of the period shift  $(T-T_0)/T$ , i.e., the part which is not attributable to just the damping through Eq. (9) but rather depends explicitly on the fluid properties. In terms of b, we have  $\Theta = 1 - \Delta + b/2 + b/2$  $O(\Delta^2, b^2)$  holding for a free disk in the case that  $\delta/R$  is of such size that the first-order terms (coefficients B) must be retained but the second-order terms (coefficients C) may still be dropped. Thus we see that, with a small but non-negligible value for b, while the period shift  $(T-T_0)/T$  and damping  $\Delta$  are nearly equal and nearly proportional to the single combination  $\rho\delta$  (i.e., proportional to  $\sqrt{\rho\eta}$ ), the higher-order terms make contributions such that the small difference between these quantities,  $(T-T_0)/T$  $-\Delta \simeq -b/2$ , is proportional to an independent combination  $\rho\delta^2$  (i.e., proportional to  $\eta$ ). This independence makes it possible to determine both  $\eta$ and  $\rho$ .

Equations analogous to Eqs. (8) for the sensitivity of  $\eta$  and  $\rho$ , as calculated simultaneously from Eqs. (7) in the case of a free disk, to fluctuations  $d\Delta$ , dT, and  $dT_0$  in the values of the measured quantities, are

$$\frac{d\eta}{\eta} \simeq \frac{2}{b} \left( \frac{-dT + dT_0}{T} + d\Delta \right)$$
(10a)

$$\frac{d\rho}{\rho} \simeq \frac{2}{b} \left( \frac{dT - dT_0}{T} - d\Delta \right).$$
(10b)

As expected, the large factor  $b^{-1}$  appears as an amplifier of errors in the measured quantities T,  $T_0$ , and  $\Delta$ .

We see from Eqs. (10) that a free oscillating-disk viscometer yields the density and the viscosity with equal precision. Actually, Eqs. (10) predict that the viscosity and density increments are exactly equal and opposite. This exact equality is a consequence of approximations made in deriving Eqs. (10); when calculated numerically from the full working equations, the viscosity and density increments differ slightly from each other (see Table I,

$\eta = 1000$ $\rho = 1000 \text{ k}$	$\mu Pa \cdot s$ $g \cdot m^{-3}$	$\delta = 1.635 \text{ mm}$ $b = B \frac{\rho R}{\bar{\rho} h} (\delta/R)^2 = 0.015$
β	2.249 mm	00
T	17.385098 s	17.641204 s
Δ	0.058796	0.053889
$\frac{b}{2\eta} \frac{\partial \eta}{\partial \Delta}$	+0.16	+1.00
$\frac{b}{2\rho}\frac{\partial\rho}{\partial\Delta}$	-0.04	-0.98
$\frac{bT}{2\eta} \frac{\partial \eta}{\partial T}$	-0.04	-0.85
$rac{bT}{2 ho} rac{\partial ho}{\partial T}$	+0.28	+1.16

Table I. Sensitivity of Simultaneously Calculated Fluid Properties<sup>a</sup>

<sup>*a*</sup> Sensitivity of the simultaneously calculated fluid properties to the period and damping for a disk between plates ( $\beta = 2.249$  mm) and for a free disk ( $\beta = \infty$ ). The instrumental parameters have the values given in Table II. The fluid properties are approximately those of water at 20°C.

where all instrumental parameters have been set to the values that pertain to our instrument). As we show below, the rough equality of viscosity and density increments remains approximately true when fixed plates are present. This situation is thus quite different from that presented by the vibrating-wire viscometer, which is also capable of simultaneous measurements of the viscosity and density [11, 12]. As we mentioned above, the vibrating-wire viscometer can determine the density with greater precision than the viscosity when a sinker is used to tension the wire.

# 3.3. Simultaneous Solutions of the Working Equations for the Disk Between Fixed Plates

Now we consider the situation when fixed plates are positioned above and below the oscillating disk at a distance  $\beta$  from the disk's flat surfaces. The behavior of the simultaneous solution of Eqs. (7) is now quite different from the free-disk case. The reason is that the  $K_1(\delta)$  and  $K_2(\delta)$  functions (Eqs. (6f) and (6g)), which attain the limits  $K_1(\delta) = 0$  and  $K_2(\delta) = 1$  when the plate spacing  $\beta$  is large compared to the boundary layer thickness  $\delta$ (i.e., in the free-disk limit), are strong, independent functions of  $\delta$  for the case where  $\beta \simeq \delta$ . The consequence is that Eqs. (7) can remain a nondegenerate system even without the contribution of the higher-order terms. The stronger independence of Eqs. (7a) and (7b) greatly increases the free part of the period shift, i.e., the part of the shift that depends explicitly on the fluid properties. As was shown in the previous sub-section, for the free disk the shift is given by  $\Theta = 1 - \Delta + b/2$ , where the free part b/2 is very small. The greater independence of T and  $\Delta$ , and thus of Eqs. (7), when fixed plates are present, can result in a significant improvement of the precision of simultaneous measurements over that achieved by the same instrument without the plates.

Because of the complexity of the  $K_1(\delta)$  and  $K_2(\delta)$  functions, the qualitative aspects of the simultaneous solution of Eqs. (7) with plates present are easier to study numerically and graphically than analytically. We therefore assign values to the instrumental parameters R,  $T_0$ ,  $\beta$ , etc., and to the measured quantities T and  $\Delta$ , and then obtain numerically solutions  $\rho$  and  $\eta$  to Eqs. (7a) and (7b), using a two-dimensional version of Newton's method. The values we assign to the instrumental parameters appear in Table II. They apply to the instrument we used for the experimental work described below and in Refs. 13 and 14. Table I shows how, for this instrument, the precision of simultaneous measurements is greatly improved by the presence of fixed plates. The table shows the case of a fluid with density  $\rho = 1000 \text{ kg} \cdot \text{m}^{-3}$  and viscosity  $\eta = 1000 \,\mu\text{Pa} \cdot \text{s}$ , values close to those of water at 20°C. (To be clear, to construct Table I, we first

 
 Table II. Instrumental Parameters of the Oscillating-Disk Viscometer

R = 33.972 mm 2h = 3.210 mm  $\beta = 2.249 \text{ mm}$   $\bar{\rho} = 8799 \text{ kg} \cdot \text{m}^{-3}$  $T_0 = 16.7894 \text{ s}$ 

solved Eqs. (7) for T and  $\Delta$  with  $\rho$  and  $\eta$  fixed at these values. Then we held T and  $\Delta$  fixed at slightly perturbed values and solved for  $\rho$  and  $\eta$ , simulating the effect of errors in the directly measured quantities on the calculated fluid properties. We did this for the case of a free disk ( $\beta = \infty$ ) and a disk between fixed plates with  $\beta = 2.249$  mm). The table shows the values of the fractional derivatives  $(\partial \eta / \partial \Delta)/\eta$ ,  $(\partial \rho / \partial \Delta)/\rho$ ,  $(\partial \eta / \partial T)(T/\eta)$ , and  $(\partial \rho / \partial T)(T/\rho)$ . Guided by Eqs. (10) we apply a factor of b/2 to these derivatives. As a result, their values are close to +1 and -1 for the free disk, in accord with the analysis of Ref. 17. For the disk between fixed plates, the magnitudes of these derivatives are much smaller, indicating a great improvement in precision.

Unfortunately, as we now show, this improvement generally is not uniform over a wide range of values of the properties of the fluid. By contrast, the sensitivity of simultaneous measurements made with a free-disk instrument depends, through b, on the density not at all and only linearly on the viscosity and thus does not vary greatly (by more than a factor of five, say) over a wide range of fluids and conditions. If the disk is between fixed plates, however, the sensitivity is likely to vary much more as the fluid properties change. In fact, we find that the plate spacing  $\beta$  and the vacuum period  $T_0$  can be chosen so that, with respect to simultaneous measurements, the disk between plates out-performs the free disk over most of the ranges of values commonly assumed by  $\rho$  and  $\eta$  in liquids. But, when the values of  $\rho$  and  $\eta$  are such that the boundary layer thickness  $\delta$  falls near one of several critical lengths  $\delta_c$ , the disk between plates in the simultaneous measurement mode becomes inferior to the free disk. These critical lengths are functions of the instrumental design. If the fluid's density and viscosity are such that  $\delta$  exactly equals one of the critical lengths, Eqs. (7) become a degenerate system and the simultaneous calculation fails.

The existence of these critical values for  $\delta$  is related to the fact that, for a fixed set of instrumental parameters and a particular assignment of the variables T and  $\Delta$ , Eqs. (7) usually have multiple simultaneous solutions  $\rho$ and  $\eta$ . Normally, these multiple roots are so well separated that there is, in practice, no ambiguity about which one actually applies to the fluid. As the fluid properties change such that the boundary layer thickness  $\delta$  approaches one of the critical lengths  $\delta_c$ , however, a pair of roots coalesces and the fractional derivatives of the type listed in Table I grow without bound. Moreover, near a critical length  $\delta_c$ , small fluctuations in the values of T and  $\Delta$  may cause Eqs. (7) to have no solution at all. By contrast, for a free disk, for any assignment  $(T, \Delta)$  for the motional parameters, Eqs. (7) always have a unique solution  $(\rho, \eta)$  for the fluid properties (provided that  $R \gg \delta$  which always holds for a practical viscometer of this type).

The physical origin of this behavior is revealed more clearly by consideration of a slightly different mathematical problem, namely the behavior of the period T and damping  $\Delta$  as we vary the plate spacing  $\beta$  at constant fluid viscosity and density. Figure 1 shows T and  $\Delta$  as functions of  $\beta$ . The calculation was done with our standard choices for the instrumental parameters (Table II) and with  $\eta = 1000 \,\mu\text{Pa} \cdot \text{s}$  and  $\rho = 1000 \,\text{kg} \cdot \text{m}^{-3}$ . With these fluid-property values and the standard value  $T_0 = 16.7894$  s for the instrument's vacuum period, the boundary layer thickness  $\delta$  has the (fixed) value of 1.635 mm. The values of T and  $\Delta$  that result for each choice for the plate spacing  $\beta$  are indicated by their fractional deviations from their free-disk values  $T_{\infty}$  and  $\Delta_{\infty}$  (see Table I). For small spacings  $\beta/\delta < 2$ ,  $\Delta$ decreases and T increases as  $\beta$  increases, while for large spacings  $\beta/\delta > 4$ , T and  $\varDelta$  are seen to saturate to their infinite-spacing values  $T_{\infty}$  and  $\varDelta_{\infty}$ . Over the intermediate range  $2 < \beta/\delta < 4$ , however, T and  $\Delta$  have nonmonotonic dependences on  $\beta$ :  $\Delta$  attains a minimum and T attains a maximum. In fact, there is an infinite series of such extremal values for each of T and  $\Delta$  as  $\beta/\delta$  increases above 2 ( $\delta$  being fixed). The amplitudes



Fig. 1. The period T and the damping  $\Delta$  as functions of the fixed-plate spacing  $\beta$ . The boundary-layer thickness  $\delta$  is fixed at 1.635 mm. The period has a maximum, and the damping has a minimum.

of these oscillations, however, decrease rapidly and only the first extremum in each series is visible in the figure. In Ref. 21, it is shown that one or the other of T and  $\Delta$  becomes extremal with respect to variation of  $\beta$  at fixed values of the fluid properties whenever  $\beta$  is approximately an integer multiple of  $\pi\delta/2^{3/2}$ , although only the first few extrema may differ appreciably from the free-disk limits. This condition has the following interpretation. The fluid velocity in the region between the fixed plates and the disk's flat surfaces, not too close to the edges, has approximately the form of a superposition of outward- and inward-traveling shear waves. The inwardtraveling wave is due to a reflection of the outward-traveling wave by the fixed plate. The relation between the wavelength  $\lambda$  and the boundary layer thickness  $\delta$  of a shear wave is  $\lambda = 2^{3/2} \pi \delta$  [22]. So, values for T and  $\Delta$ extremal with respect to variation of  $\beta$  occur when the plate spacing is an integral multiple of  $\lambda/8$ . For such spacings, at the surface of the disk the phase of the returning reflected wave, which has traveled a total distance of 2 $\beta$ , differs from the phase of the outgoing wave by 0,  $\pi$ , or  $\pm \pi/2$ . Each extremal condition (maximum or minimum of T or  $\Delta$ ) corresponds to one of these four phase differences.

Returning to the problem of the simultaneous calculation of viscosity and density, we reiterate that the critical condition for the method's failure is that two normally distinct pairs of fluid property values, say  $(\rho_1, \eta_1)$  and  $(\rho_2, \eta_2)$ , which solve Eqs. (7) as functions of the motional parameters  $(T, \Delta)$ , coalesce into a single, doubly degenerate solution  $(\rho, \eta)$  for some T and  $\Delta$ . The existence of multiple solutions to Eqs. (7) and the possibility of their coalescence is again due to phase-interference effects between the outward-traveling shear wave and its inward-traveling reflection. Unlike the behavior of T and  $\Delta$  considered as functions of  $\beta$  at constant  $\rho$  and  $\eta$ , however, the critical condition for the simultaneous measurement of  $\rho$  and  $\eta$  as functions of T and  $\Delta$  at constant  $\beta$  does not give rise to extremal values for any quantities, nor is the phase relation between the outwardand inward-traveling shear waves particularly simple when the critical condition holds. In Ref. 21 it is shown that the critical condition holds when the fluid properties (and the instrument's vacuum period  $T_0$ ) are such that the boundary layer thickness  $\delta$  takes on one of a discrete set of critical lengths  $\delta_c$  which depend only on R, h, and  $\beta$ . To find these critical lengths as functions of R, h, and  $\beta$ , it is generally necessary to proceed numerically according to a recipe given in Ref. 21. Taking R, h, and  $\beta$  as specified in Table II, we find that our instrument has two critical lengths:  $\delta_c = 0.495$ and 0.753 mm. The second of these lengths can be important when our instrument is used in the simultaneous mode. For example, water near 140°C has properties  $\rho \simeq 920 \text{ kg} \cdot \text{m}^{-3}$  and  $\eta \simeq 180 \,\mu\text{Pa} \cdot \text{s}$  which, with our instrument's vacuum period  $T_0 = 16.7894$  s, yield  $\delta \simeq 0.72$  mm for the boundary layer thickness. The existence of our instrument's smaller critical length is less likely to be important, since a typical liquid would seldom be of such low viscosity and high density as to yield  $\delta$  near the smaller critical length.

#### 3.4. Approximate Analytic Treatment of the Disk Between Fixed Plates

An analytic treatment of simultaneous calculations, even if applicable only in a special case, is a much desired supplement to numerical work, both as a guide in determining an instrument's critical lengths and for the derivation of formulas analogous to Eqs. (10). An approximate analytic treatment is possible in the special case of disks with high aspect ratio  $(R \gg h)$  for which  $R \gg \delta$  holds so strongly that the higher-order terms in Eqs. (7) can be dropped [21]. For brevity, we call this the case of a flatsdominated disk, since in this limit one need take account only of the drag on the flat surfaces of the disk and the edge effect may be ignored. (As has been discussed above, simultaneous measurements fail in this limit if the disk is free, for the assumption is equivalent to setting b = 0.) For the flatsdominated disk, only the terms in  $K_1(\delta)$  and  $K_2(\delta)$  survive on the righthand sides of Eqs. (7). Also, the damping and period shift are small in this limit, so  $H_1 \simeq H_2 \simeq 1/\sqrt{2}$  are good approximations and the arguments of all the transcendental functions that appear in the definitions of  $K_1(\delta)$  and  $K_2(\delta)$  reduce to a common variable  $z = \sqrt{2} \beta/\delta$  (see Eqs. (6)). The condition for the coalescence of double solutions of Eqs. (7), according to which  $\delta$  is approaching one of the instrument's critical lengths  $\delta_{c}$ , then becomes a transcendental equation in the variable z. Only one root exists to this equation, implying that there exists a unique critical length whose value  $\delta_{\rm c} = 0.36016\beta$  depends only on the fixed plate spacing  $\beta$ . The critical condition for the breakdown of simultaneous measurements is therefore  $\delta = \delta_c = 0.36016\beta$ , which, as noted above, does not imply a simple ratio between the wavelength of the shear wave and the plate spacing. However, the existence of multiple solutions and their possible coalescence is due to interference effects between the outward- and inward-going shear waves, for it is just these phase effects that are described by the  $K_1(\delta)$  and  $K_2(\delta)$ functions.

The finding  $\delta_c = 0.36016\beta$  for the flats-dominated disk between fixed plates can be tested by applying it to our instrument, for which  $R \gg h$  and  $R \gg \delta$  hold rather well. Our instrument's two critical lengths,  $\delta_c = 0.495$  and 0.753 mm, can be written, respectively, as  $\delta_c = 0.220\beta$  and 0.335 $\beta$ , where  $\beta = 2.249$  mm is the fixed plate spacing. (Recall, though, that both these critical lengths depend on the disk's radius *R* and half-height *h*, which

have been set to the values appearing in Table II for the numerical computation of the critical lengths  $\delta_{c}$ .) The rough agreement of the coefficients 0.335 and 0.36016 shows that our instrument's larger critical length corresponds to the unique critical length of the ideal flats-dominated disk between plates, but the exact value for the length is modified by the small but finite ratios h/R and  $\delta/R$  that hold for our instrument. The instrument's smaller critical length, on the other hand, owes its very existence to these non-vanishing ratios, and no account of it is given by the treatment of the ideal flats-dominated disk. We can expect that any instrument that maintains  $R \gg h$  and  $R \gg \delta$  has a critical length near one third of the fixed plate spacing  $\beta$ . Its other critical lengths, if any, depend on R, h, and  $\beta$  in an unknown way and have to be searched for numerically. Which, if any, of these critical lengths impacts on a program of simultaneous measurements depends on the instrument's vacuum period  $T_0$ : it may be possible to choose  $T_0$  such that the boundary layer thickness  $\delta$  stays well away from a critical length  $\delta_c$  over the ranges of viscosity and density that are of interest. This is true in the case of our instrument's smaller critical length, but, as noted above, its larger critical length  $\delta_c = 0.335\beta$  can affect simultaneous measurements when the viscosity is low and the density is high, relative, say, to a typical organic liquid.

#### 3.5. Consequences for Precision of Simultaneous Measurements

The analytic treatment of the flats-dominated disk between fixed plates also yields equations analogous to Eqs. (10) for the precision of simultaneous measurements:

$$\frac{d\eta}{\eta} \simeq 0.2 \, \frac{\bar{\rho}h\beta}{\rho\delta^2} \left( \frac{-dT/T + dT_0/T + d\Delta}{\delta/\beta - \delta_c/\beta} \right),\tag{11a}$$

$$\frac{d\rho}{\rho} \simeq 0.2 \, \frac{\bar{\rho}h\beta}{\rho\delta^2} \left( \frac{dT/T - dT_0/T - d\Delta}{\delta/\beta - \delta_c/\beta} \right),\tag{11b}$$

where  $\delta_c = 0.36016\beta$ . Comparison of Eqs. (11) and (10) shows that simultaneous measurements made with a disk between plates will tend, overall, to be more precise than those made with a free disk, because the prefactor  $0.2\bar{\rho}h\beta/(\rho\delta^2)$  may be considerably smaller than the factor  $2/b = 2\bar{\rho}hR/(B\rho\delta^2)$  (by a factor of about 0.02 in the case of our instrument). In Eqs. (11), however, an amplifying factor  $\beta/(\delta-\delta_c)$  diminishes this gain and, with  $\delta$  sufficiently close to  $\delta_c$ , results in the failure of the simultaneous method. Equations (11) are only rough approximations. For example, like Eqs. (10) for the free disk, they predict that the viscosity and density increments are equal and opposite, but reference to Table I, which shows the increments calculated exactly from the full form of Eqs. (7), shows that this prediction is obeyed only crudely.

We conclude this section with a graphical illustration of the main points. Figure 2 illustrates how the fractional viscosity increment  $d\eta/\eta$ due to fixed increments  $d\Delta$  and dT changes as the fluid properties take on a range of values. To make the plot, we held the density fixed at  $\rho =$ 1000 kg·m<sup>-3</sup> and solved Eqs. (7) for T and  $\Delta$  as functions of the viscosity. Then we applied to these values of T and  $\Delta$  the increments dT = +0.1 ms and  $d\Delta = +4 \times 10^{-6}$ , which are representative of the precision with which the period and damping are measured in our laboratory. Finally, we resolved Eqs. (7) for  $\eta$  and  $\rho$  with T and  $\Delta$  assigned their incremented values. This process imitates the introduction of errors into the measurements of the period and damping. Two such calculations were carried out, one for the free disk and one for the disk between plates with  $\beta = 2.249$  mm, with all other instrumental parameters as given in Table II.

For all viscosities slightly above 200  $\mu$ Pa·s, the viscosity increment obtained when plates are present is much smaller than the increment obtained for a free disk, indicating that a more precise measurement results when the disk is between fixed plates. Around 200  $\mu$ Pa·s, however, the viscosity increment with plates present becomes very large as the boundary layer thickness  $\delta$  comes close to the instrument's critical length  $\delta_c =$ 0.735 mm. The broken curves show the results of the two calculations of the viscosity increment made as described, i.e., by direct, numerical solutions of Eqs. (7) in their full form. The smooth curve was calculated from

Fig. 2. The relative error  $d\eta/\eta$  of the viscosity when it is measured simultaneously with the density. The density  $\rho$  is fixed at 1000 kg·m<sup>-3</sup> and the error is shown as functions of the viscosity  $\eta$  and of the boundary-layer thickness  $\delta = (\eta T_0/2\pi\rho)^{1/2}$ . Fixed errors dT = 0.1 ms and  $d\Delta = 4 \times 10^{-6}$  are assumed for the period and damping. The broken curves have been calculated numerically from the working equations Eqs. (7). The smooth curve is a plot of Eq. (11a), an approximation derived by taking account only of the drag on the disk's flat surfaces.



Eq. (11a), which applies to flats-dominated disks. In using Eq. (11a), however, we set  $\delta_c = 0.735$  mm, which is one of our instrument's actual critical lengths as determined by the numerical treatment of Eqs. (7) in their full form, rather than to the value  $\delta_c = 0.36016\beta = 0.810$  mm predicted by the approximate analytical treatment of the flats-dominated disk.

Since these values for the critical length are not too different and since the right branches of the two curves describing the disk between plates lie very close, we can conclude that simultaneous measurements made with our instrument are given a good qualitative description by the flats-dominated disk approximation, provided the fluids studied have properties such that the boundary-layer thickness is larger than, say,  $\delta = 0.7 \text{ mm}$  (see Fig. 2). This will be the case for most liquids. For such fluids, the flatsdominated disk approximation gives a rough estimate of the precision that can be attained, including the loss of all precision that occurs when  $\delta$ approaches  $\delta_c = 0.735$  mm. For fluids such that  $\delta < 0.7$  mm, on the other hand, the exact and approximate increments  $d\eta/\eta$  disagree greatly because, as noted above, the flats-dominated disk has only one critical length. The actual instrument, however, has finite values for the ratios h/R and  $\delta/R$ which engender a second critical length, in this case at  $\delta_c = 0.495 \text{ mm}$ , where the fluctuations of the calculated fluid properties again become infinite. The effects of this second divergence can be seen in the figure; they are not accounted for by the flats-dominated disk approximation. Therefore, it would be well to determine all of the critical lengths of one's instrument numerically, and apply Eqs. (11) with care.

# 4. EXPERIMENT

A complete description of our oscillating-disk viscometer can be found in a previous publication [14]. Here we mention only a few details. Because the edge effect in the case of comparable fixed plate spacing and boundary layer thickness is unknown, we use in the working equations the expression for the edge effect of a free disk (i.e., Eq. (5)), and then remove the resulting error by means of a calibration [13, 14]. To calibrate Eqs. (7), we fill the viscometer with water, whose properties we take from the literature [23, 24]; then we measure the period T and the damping  $\Delta$ . Next we evaluate the right and left sides of Eqs. (7), generally finding that they are not equal. We make them equal by adding within the braces of the right sides of Eqs. (7a) and (7b), respectively, the required numbers F and E. We carry this out over a range of temperature so that we may then express E and F as polynomials in  $\delta$ . These calibration functions  $E(\delta)$  and  $F(\delta)$  are then incorporated into Eqs. (7) for measurements on fluids with unknown properties. We find that  $E(\delta)$  and  $F(\delta)$  lie within  $\pm 0.01$ , whereas they add to terms in Eqs. (7) that are of order unity. Consequently, they do not affect the qualitative behavior of Eqs. (7) that is our main concern. They do, however, cause small shifts in the values of the instrument's critical lengths. The calibrated working equations have a critical length at  $\delta_c = 0.750$  mm instead of 0.735 mm.

Our instrument has a metallic suspension wire made from a platinum/ tungsten alloy selected for minimum hysteresis of its elastic properties under temperature and pressure cycling, and for optimal reproducibility of the rest position of the disk. The stiffness of the wire depends on temperature, so it is necessary to take account of the variation of the vacuum period  $T_0$  with temperature. In fact, the vacuum period  $T_0$  is better defined as the period that would be measured in the absence of the hydrodynamic drag exerted by the fluid, but under the otherwise identical conditions that apply during the measurement with the fluid present. Since the moment of inertia of the disk depends on the pressure (i.e., because of compression), it is also necessary to account for the variation of  $T_0$  with pressure. Table II gives the value of  $T_0$  at 25°C and zero pressure. The temperature and pressure coefficients of  $T_0$  are approximately 2.1 ms  $\cdot$  K<sup>-1</sup> and -0.15 ms  $\cdot$  MPa<sup>-1</sup> [13]. It may be noted that although  $T_0$  applies in vacuo, its value derives from measurements made with fluid present. To obtain  $T_0$  and its temperature dependence, we measure the period in a gas at atmospheric pressure, then apply a correction for the drag exerted on the disk by the gas [17, 25]. The pressure coefficient of  $T_0$  is inferred from the period in pressurized water [13].

We have used the instrument to measure the viscosity of liquid toluene over the temperature and pressure ranges of 25 to 150°C and 0 to 30 MPa [14]. Figure 3, reproduced from Ref. 14, gives an overview of these data. In Ref. 14, we gave the viscosities as we computed them from the damping equation (Eq. (7b)) with the density of toluene calculated from a published equation of state [15]. Here (as in Ref. 13), we re-analyze the period and damping data to give the viscosity and density simultaneously. Results for a typical isobar, P = 15.0 MPa, are shown in Fig. 4 in the form of deviation plots of the simultaneously calculated viscosity from the values published in Ref. 14 and the simultaneously calculated density from the values predicted by the equation of state published in Ref. 15. (These plots also appeared in Ref. 13.) For both properties the deviations fall within  $\pm 0.4\%$ for all pressures and for temperatures up to 125°C. Deviations a little larger than  $\pm 1\%$  occurred for the highest temperature, 150°C. We attribute these larger deviations to inaccuracies of our temperature measurements at the highest temperatures, which cause inaccuracy in the calculation of the density from the equation of state. (The equation of state may itself be in appreciable error at the higher temperatures: the measurements of Ref. 15



Fig. 3. Viscosity of toluene, from Ref. 14.
○, 24°C; ●, 30°C; ▽, 41°C; ♥, 49°C;
□, 60°C; ■, 77°C; △, 98°C; ▲, 121°C;
◇, 152°C.

upon which it is based were made at temperatures that did not exceed 100°C. To treat the higher temperatures, we simply extrapolated the density equation of Kashiwagi et al. [15].) The precision of the simultaneous measurements, estimated from the scatter of the values found from three measurements made at each temperature and pressure (and which have been averaged in the figure), was always at the level of  $\pm 0.2\%$  or better. This level of precision is about the same as that which we achieved when the damping equation was used alone with the density specified. Thus, in these simultaneous measurements we were not affected by the phenomenon discussed above, namely the sharp loss of precision that occurs over certain restricted ranges of properties of the fluid. The reason is



Fig. 4. Viscosity and density of toluene obtained by simultaneous solution of the coupled working equations Eqs. (7), as functions of temperature. The viscosities are compared to those we reported in Ref. 14, where we used an equation of state for the density [15] and solved only the damping working equation. The densities are compared to the predictions of the equation of state reported in Ref. 15. Data shown are for the P = 15.0 MPa isobar.

simply that in these measurements with toluene, the boundary layer thickness  $\delta$  never fell below 0.84 mm and therefore never approached the instrument's critical length  $\delta_c = 0.75$  mm.

We may conclude that for measurements of viscosity with our oscillating-disk viscometer, a knowledge of the fluid density is not necessary because, by using all the information contained in the measurements of the period and damping, and the complex equation that relates these quantities to the fluid properties, it is possible to obtain results essentially identical to those we obtain by the conventional method in which the density is supplied independently. Specifically, with our instrument, a typical precision of the simultaneously measured viscosity is  $\pm 0.2\%$ , while the agreement of the simultaneously measured viscosity with the viscosity obtained from the damping equation alone is typically  $\pm 0.4\%$ . Moreover, in the simultaneous method along with the viscosity we can measure the density with a typical precision of  $\pm 0.2\%$  and an uncertainty of  $\pm 0.4\%$ .

New correlations for the density and viscosity of toluene have been recommended recently by Assael et al. [26]. Figure 5 compares the viscosity we found by the simultaneous method with the values predicted by the correlation of Ref. 26. The 15.0 MPa isobar is again shown as typical, but only the temperatures below 100°C are shown since this temperature is the limit of the correlation's validity. The deviations are between 0 and +2%. The claimed accuracy of the viscosity correlation is  $\pm 2\%$ . The simultaneously determined density also can be compared with the equation of state recommended by Assael et al. [26]. Over their ranges of validity (temperatures up to 100°C), the density equations of Refs. 15 and 26 agree within  $\pm 0.1\%$ , and their extrapolations to 150°C agree within  $\pm 0.2\%$ . Therefore, this comparison results in a plot very similar to Fig. 4b.

#### 5. CONCLUSIONS

In this paper we have considered the use of oscillating-disk viscometers for simultaneous measurements of the viscosity and density of a fluid. We

Fig. 5. Viscosity of toluene obtained by simultaneous solution of the coupled working equations Eqs. (7), as functions of temperature. The viscosities are compared to a recently recommended correlation [26]. The pressure is 15.0 MPa; the temperature is restricted to the correlation's range of validity.



Krall and Sengers

found that a properly designed instrument is capable of yielding for the viscosity essentially the same results that are obtained by the more commonly used method in which the density is supplied independently. This procedure allows the measurement of the viscosity of fluids of unknown density and, in the process, determines the density at the same level of precision that is attained for the viscosity. This precision is typically at the level of several tenths of a percent, however, so with regard to density the method is out-performed by many stand-alone densimeters.

As a practical demonstration of the method, we re-analyzed in the simultaneous calculation mode some raw data we previously collected for a study of the viscosity of liquid toluene [14]. We found that with the re-analysis according to the simultaneous mode of calculation, we essentially reproduced our previous viscosity values, which had been found by the conventional analysis in which the density is independently supplied. The agreement was within  $\pm 0.4\%$  except at the very highest temperatures. The new analysis also furnished measurements of the density with the same level of accuracy.

We discussed at length a subtle effect, not commented on before, which is introduced into the simultaneous method by the presence of the fixed plates often used with oscillating-disk viscometers. Fixed plates tend to improve greatly the precision of simultaneous measurements. At the same time, however, it has to be recognized that the presence of fixed plates is responsible for the existence of multiple solutions of the working equations, and causes the precision of simultaneous measurements to fall dramatically when the ratio of the viscosity to the density falls close to certain values that depend on the instrument's design. Also, for simultaneous measurements it is necessary to know the disk's period of oscillation in vacuo  $T_0$  with as much accuracy as is required for the measurement of T, the period with fluid present. This requirement makes a significant demand on the instrument's stability and also requires the experimenters to take account of small effects, such as the dependence of  $T_0$  on pressure, which are not important when the viscosity is calculated from the damping with the density supplied independently.

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