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Shear Thinning in Xenon

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Summary

We measured shear thinning, a viscosity decrease ordinarily associated with complex liquids such as molten plastics or ketchup, near the critical point of xenon. The data span a wide range of dimensionless shear rate: $10^{-3} < \dot{\gamma}\tau < 700$, where $\dot{\gamma}$ is the shear rate and τ is the relaxation time of critical fluctuations. As predicted by theory, shear thinning occurred when $\dot{\gamma}\tau > 1$. The measurements were conducted aboard the *Space Shuttle Columbia* to avoid the density stratification caused by Earth's gravity.

Introduction

Both oil in a car engine and paint on a paintbrush need viscosity control. Sliding engine parts sometimes shear the intervening oil layer fast enough to decrease the viscosity of the oil's polymer additives, and such shear thinning is bad for the engine. Conversely, brushing a well-engineered paint onto a wall temporarily decreases the paint's viscosity, and shear thinning helps to spread the paint. In general, shearing any fluid fast enough to distort its microscopic structure will change the viscosity. Shear thinning, a decrease in viscosity with increasing shear rate, is common in complex fluids from molten plastics to ketchup; such fluids have microscopic structures that relax slowly in comparison with the shear rate $\dot{\gamma}$. Here, we report the first observation of shear thinning in a simple fluid. A viscometer measured the drag on a delicate nickel screen as it oscillated in a sample of xenon at its critical density. Upon approaching the liquid-vapor critical point (Fig. 1), the relaxation time τ of the critical fluctuations increased by orders of magnitude, and shear thinning occurred when $\dot{\gamma}\tau > 1$.

Predictions of shear thinning usually rely on a detailed model of the molecules and their interactions. In contrast, the theory for viscosity near critical points (Refs. 1 to 10) assumes only that the interactions are short ranged and that the viscosity is dominated by the microscopic fluctuations that occur in all fluids. Because of this generality, an understanding of how distorted fluctuations cause shear thinning may lead to general insights for rheology, which describes the dependence of viscosity on shear rate and frequency.

The critical fluctuations have a size ξ that increases as a fluid approaches its liquid-vapor critical point. Coupling between fluctuations of density and velocity causes the viscosity η to increase as the power law $\eta \propto \xi^{x_\eta}$. The viscosity increase is small because the universal exponent $x_\eta = 0.069$ is small (Refs. 10 and 11). However, τ has a much larger increase because it is nearly proportional to the volume of a fluctuation ξ^3. More precisely, τ varies as follows (Ref. 3):

$$\tau = \tau_0 \left(\frac{T - T_c}{T_c} \right)^{-1.93} \propto \xi^{3.069} \tag{1}$$

which increases as the temperature T approaches the critical temperature T_c. Xenon at its critical density ($\rho_c = 1.1$ g/cm^3) is a monatomic fluid with a convenient critical temperature of $T_c = 17$ °C. In Equation (1), the time-constant amplitude τ_0 is tiny (1 ps), but we achieved a relaxation time as large as $\tau = 1$ s during the microgravity measurements at 0.19 mK above the critical temperature.

Shear thinning occurs when $\dot{\gamma}$ exceeds $1/\tau$. A related phenomenon, viscoelasticity, occurs at small shear rates when the shearing is oscillatory at a frequency f that exceeds $1/\tau$. The first accurate measurement of near-critical viscoelasticity was made by a previous microgravity experiment, Critical Viscosity of Xenon (CVX, Ref. 11). The present experiment, CVX–2, measured shear thinning by driving the same viscometer to amplitudes 30 times larger. This report summarizes the key results of the CVX–2 experiment. The interested reader can find a detailed description of the experiment and the data analysis in Reference 12. A list of symbols used in this report is presented in the appendix.

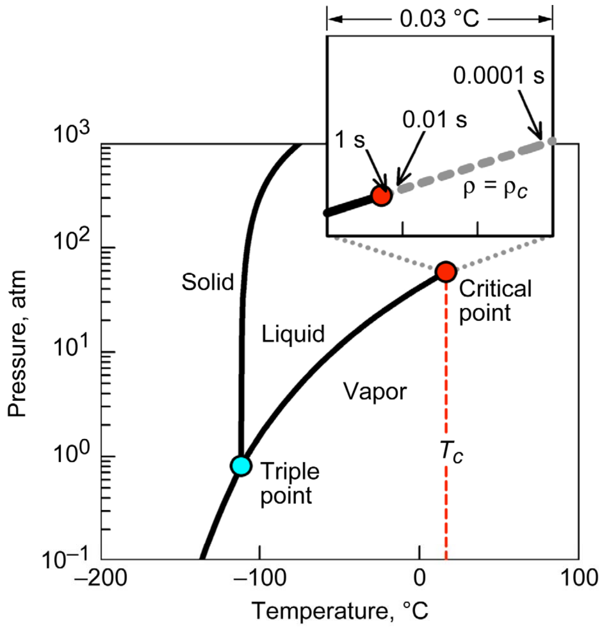

Figure 1.—Xenon phase diagram. Because the experiment was conducted in microgravity, the region near the critical point became accessible, thus making it possible to measure shear thinning. The inset shows how the fluctuation relaxation time τ increased along the path of critical density used for the measurements.

In general, the viscosity $\eta(f\tau, \dot{\gamma}\tau)$ depends on both the dimensionless frequency $f\tau$ and dimensionless shear rate $\dot{\gamma}\tau$, but so far theory only handles viscoelasticity in the limit of zero shear rate, $\eta(f\tau,0)$, and shear thinning in the limit of zero frequency, $\eta(0,\dot{\gamma}\tau)$. Therefore, we analyzed our data by approximating xenon's constitutive relation by the product of those two limiting cases:

$$\eta(f\tau, \dot{\gamma}\tau) \simeq \eta(f\tau,0)\frac{\eta(0,\dot{\gamma}\tau)}{\eta(0,0)} \quad (2)$$

The viscoelastic behavior $\eta(f\tau,0)$ is known from theory (Ref. 6) and has been confirmed by experiment (Ref. 11). The shear-thinning behavior,

$$\frac{\eta(0,\dot{\gamma}\tau)}{\eta(0,0)} = \left[1 + A_\gamma|\dot{\gamma}\tau|\right]^{-p} \quad (3)$$

with $p = x_\eta/(3 + x_\eta) = 0.022$, is based on an analogy between near-critical fluids and polymer fluids: Douglas (Ref. 13) pointed out that the empirical Carreau-Yasuda relation (Ref. 14) used for many polymer fluids is consistent with the results from the renormalization group theory for near-critical fluids in the limits of small and large shear rates.

The shear-thinning function (Eq. (3)) also works at intermediate shear rates, but theory and experiment had disagreed about the value of the shear-rate scale factor A_γ (Ref. 15). The value $A_\gamma = 0.121$, which describes Oxtoby's numerical results from mode-coupling theory (Ref. 1), is twice the value from Hamano et al.'s measurements, $A_\gamma = 0.067 \pm 0.007$ (Ref. 16). Like all previous measurements of near-critical shear thinning, Hamano et al. measured a binary liquid mixture instead of a monatomic fluid. However, their results are particularly valuable because they used a Couette viscometer to create a uniform shear field.

Experiment and Analysis

The density of a fluid near its liquid-vapor critical point is enormously sensitive to gradients of pressure and temperature. Shear thinning cannot be observed in quiescent xenon on Earth because the gradient of pressure caused by Earth's gravity stratifies the density so that the layer near the critical density is too thin to be studied in a viscometer. To reduce gravitational stratification, the measurements were conducted in microgravity aboard *Space Shuttle Columbia* in 2003 during the ill-fated mission STS–107. Much of the data was downlinked during the flight, and the unexpected recovery of the hard disk drive from *Columbia*'s debris made 99 % of the data available.

To limit temperature gradients, the 11-cm^3 xenon sample was sealed into a thick-walled copper cell (Fig. 2) that was surrounded by three temperature-controlled aluminum shells. That arrangement limited temporal variations of temperature to 10 μK and spatial variations to less than 0.2 μK. The viscosity was determined by measuring the drag on an oscillating rectangle of nickel screen. Its small mass (1 mg) made it insensitive to vibrations of the space shuttle's structure. One of the screen wires was extended out of the rectangle and soldered to a stiff yoke to act as a torsion spring. The yoke supported the screen between four electrodes placed parallel to the screen.

Oscillating voltages applied to the electrodes generated a calculable torque on the screen that caused it to oscillate much like a child's seesaw at frequencies of $f = 1$, 2, 3, or 5 Hz. The applied torque plus the opposing hydrodynamic torque determined the oscillating screen's angular displacement, which was detected by the unbalance of a capacitance bridge. We obtained the hydrodynamic torque from the ratio of the Fourier transforms of torque and displacement. See Reference 11 for more details about the viscometer.

Attention was focused on the wires with the largest shear rate, specifically the wires located at both tips of the oscillator. Because the angular displacements were less than 0.05 radians, the tips' motion was approximated as a linear displacement of amplitude x_0. The hydrodynamic drag force on the tip wire, $F(x_0, f, \dot{\gamma}\tau)$, which depended on x_0 and f as well as on the dimensionless shear rate $\dot{\gamma}\tau$, was deduced from the hydrodynamic torque on the entire oscillator and its derivative with respect to amplitude.

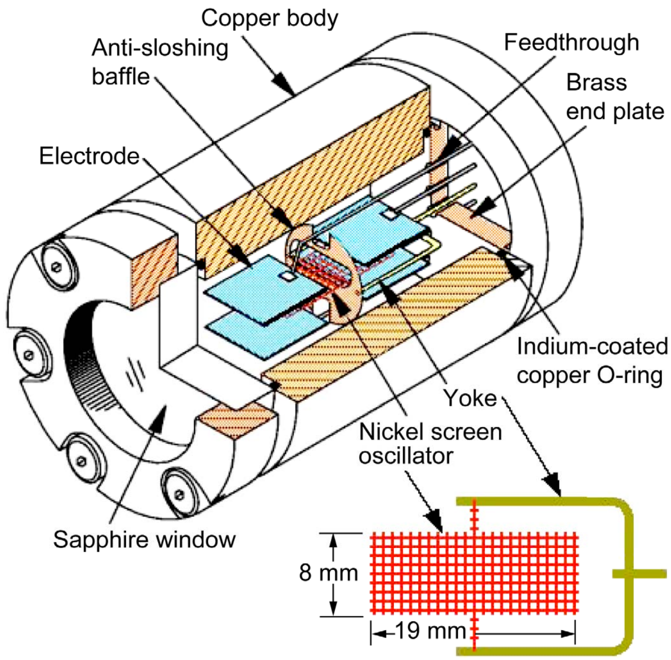

Figure 2.—Cutaway view of the viscometer cell. Applying opposite oscillating voltages to diagonally opposite pairs of electrodes caused the grounded screen to oscillate about its torsion axis. Adapted from Reference 12; prepared for NASA.

The qualitative signature of shear thinning near T_c was a nonlinear dependence of the drag force F as a function of the oscillator's amplitude x_0; the drag decreased as the amplitude increased. However, this signature was superimposed on an increase in the drag due to the convective term of the Navier-Stokes equation $[\rho(\nu \bullet \nabla)\nu]$, which is not related to critical fluctuations. Therefore, we measured just the convective contribution to F in the Newtonian fluid far from T_c and extrapolated its value into the non-Newtonian region near T_c. Dividing each non-Newtonian value of F by the extrapolated Newtonian value removed the Navier-Stokes nonlinearity and its noncritical dependence on x_0. Additional measurements made at a small amplitude x_{small} were used to remove the effect of viscoelasticity and its dependence on f. The resulting force ratio,

$$C_\gamma(\dot{\gamma}\tau) = \frac{\left[\dfrac{F(x_0, f, \dot{\gamma}\tau)}{F(x_{\text{small}}, f, 0)}\right]_{\text{non-Newtonian}}}{\left[\dfrac{F(x_0, f, 0)}{F(x_{\text{small}}, f, 0)}\right]_{\text{extrapolated Newtonian}}} \qquad (4)$$

was assumed to depend only on the dimensionless shear rate $\dot{\gamma}\tau$.

Results and Discussion

Figure 3 shows the measured magnitude and phase of the force ratio $C_\gamma(\dot{\gamma}\tau)$. The shear rate varies by a factor of 1.8 or more, and the frequency varies by a factor of 5, while as predicted by theory, the data vary only with dimensionless shear rate. At large dimensionless shear rates, the force decreased by as much as 1 % below the force in the absence of shear thinning. Although small, the decrease is remarkable because it indicates shear thinning of a monatomic fluid.

Figure 3 also shows the results from computational fluid dynamics (CFD) calculations, which had two purposes. The first purpose was to estimate the shear rate at the surface of the experimental oscillator. The second purpose was to estimate the force ratio $C_\gamma(\dot{\gamma}\tau)$ in non-Newtonian xenon. To do so, the CFD calculations used Equation (3) with the value $A_\gamma \equiv 0.067$, which describes Hamano et al.'s experimental data. Viscoelasticity was ignored because the assumed constitutive relation of Equation (2) causes viscoelasticity to cancel out of the force ratio of Equation (4).

The calculations yielded the force on a circular cylinder oscillating perpendicular to its axis. The fluid density and viscosity were matched to the experimental xenon, and the cylinder diameter was matched to the effective hydrodynamic diameter of the screen wires (Ref. 11), which had a roughly rectangular cross section of 8 by 30 μm. The circular cross section used by the CFD calculations allowed a direct check against Stokes' analytical result (Refs. 17 and 18) at small amplitudes. At both small and large amplitudes, the CFD calculations approximated the response of the experimental oscillator in the Newtonian xenon far from T_c.

There is no accurate analytical expression for $C_\gamma(\dot{\gamma}\tau)$, the force ratio of a cylinder oscillating in a shear-thinning fluid. Therefore, we derived an approximate expression for the real quantity $1-\left|C_\gamma(\dot{\gamma}\tau)\right|$ and generalized it with two free parameters to obtain the fitting function

$$1 - C_\gamma^{\text{magFit}}(\dot{\gamma}\tau) \equiv \alpha_{\text{mag}}\left\{1 - \left[1 + \left(\frac{\beta_{\text{mag}}}{\alpha_{\text{mag}}}\right)\dot{\gamma}\tau\right]^{-p}\right\} \qquad (5)$$

At large shear rates, the empirical parameter α_{mag} allows for errors due to the approximate nature of Equation (5) (Ref. 12). At small shear rates, Equation (5) reduces to the simple expression

$$1 - C_\gamma^{\text{magFit}}(\dot{\gamma}\tau) \simeq p\,\beta_{\text{mag}}\dot{\gamma}\tau \qquad (6)$$

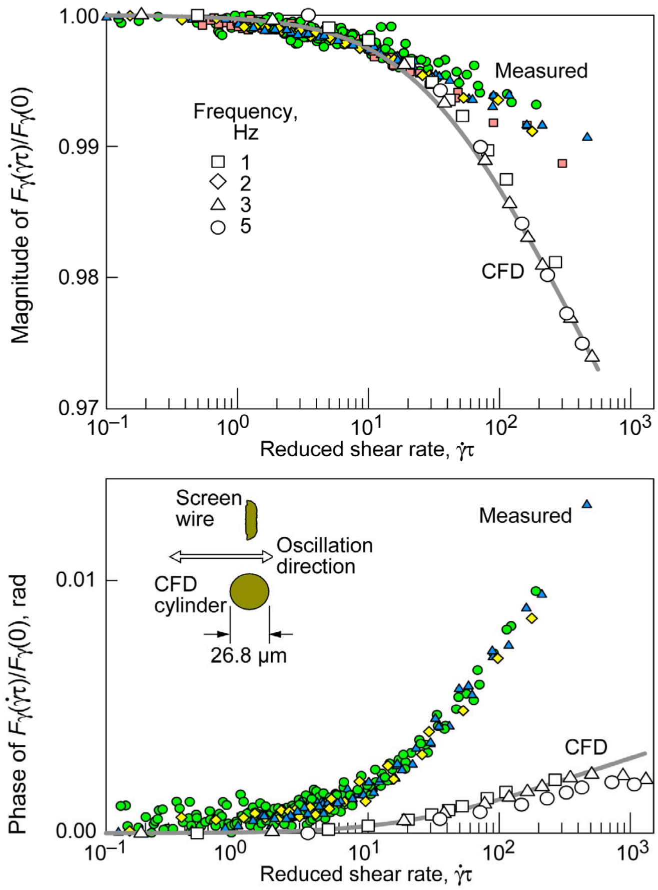

Figure 3.—Magnitude and phase of the hydrodynamic force ratio $C_\gamma(\dot{\gamma}\tau)$ on the tip wire of the viscometer (Eq. (4)). The data are normalized by the force expected in the absence of shear thinning, and the deviations from the values at $\dot{\gamma}\tau < 1$ indicate shear thinning. (The experimental 1-Hz phase data had unexpected noise and are not shown.) The computational fluid dynamics (CFD) calculations used a circular cross section that differed from that of the screen wire, and they assumed that viscoelasticity canceled out of the force ratio. Adapted from Reference 12; prepared for NASA.

Equation (6) has the same form as Equation (3) in the limit of small shear rate; its only free parameter β_{mag} is thus directly sensitive to the shear-rate scale factor A_γ.

Equation (5) was fit to values of $1-\left|C_\gamma\left(\dot{\gamma}\tau\right)\right|$ for both the CFD data sets and the experimental data sets. Figure 4 shows that the fitted values of β_{mag} have little frequency dependence but are larger than the values fit to the CFD results. The ratio

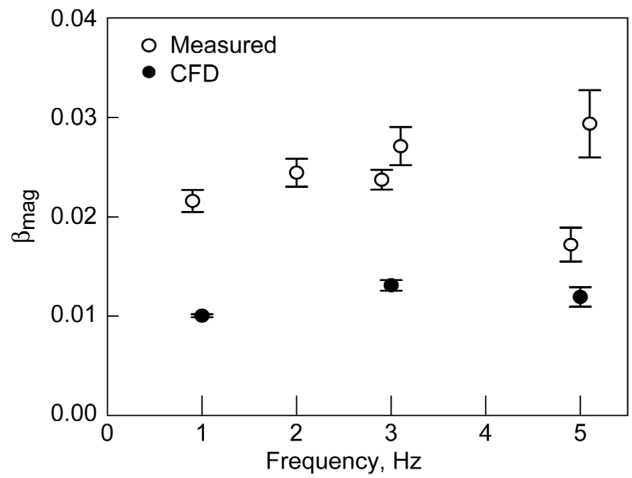

Figure 4.—Results of fitting Equation (5) to the magnitude of the force ratio C_γ. (Small horizontal displacements were added for clarity.) Both the measured values and the CFD values of β_{mag} have little frequency dependence, but the measured values are larger. Thus, the measured shear-rate scale factor A_γ was larger than the value assumed for the CFD calculations. Adapted from Reference 12; prepared for NASA.

between the measured and CFD values of β_{mag} yields the result for A_γ in Equation (3):

$$A_\gamma = 0.137 \pm 0.029 \tag{7}$$

This value, obtained at the liquid-vapor critical point of xenon, agrees with Oxtoby's mode-coupling result of 0.121. A remaining challenge is to explain the disagreement between the computational fluid dynamics and experimental results at the large shear rates shown in Figure 4. Further measurements as well as theoretical work will be needed before Equation (2) can be replaced by a constitutive equation that is accurate at large shear rates.

Concluding Remarks

Shear thinning, a viscosity decrease common in complex liquids, was observed for the first time in a simple monatomic fluid. The measurements were made possible by conducting the Critical Viscosity of Xenon – 2 experiment on the *Space Shuttle Columbia*. The measured shear-rate scale factor of 0.137±0.029 is in agreement with Oxtoby's mode-coupling result of 0.121.

Appendix—Symbols

A_γ	shear-rate scale factor
$C_\gamma(\dot{\gamma}\tau)$	shear-thinning force ratio
$C_\gamma^{\mathrm{magFit}}(\dot{\gamma}\tau)$	emprical analytical representation of the real part of $C_\gamma(\dot{\gamma}\tau)$
F	hydrodynamic drag force
f	frequency
$f\tau$	dimensionless frequency
T	temperature
T_c	critical temperature
x_n	universal critical exponent for viscosity
x_{small}	small amplitude oscillation
x_0	linear tip wire displacement
α_{mag}	empirical fitting parameter
β_{mag}	empirical fitting parameter
$\dot{\gamma}$	shear rate
η	viscosity
ρ_c	critical density
τ	relaxation time of critical fluctuations
τ_0	time-constant amplitude
ξ	fluctuation length

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14. ABSTRACT
We measured shear thinning, a viscosity decrease ordinarily associated with complex liquids such as molten plastics or ketchup, near the critical point of xenon. The data span a wide range of dimensionless shear rate: the product of the shear rate and the relaxation time of critical fluctuations was greater than 0.001 and was less than 700. As predicted by theory, shear thinning occurred when this product was greater than 1. The measurements were conducted aboard the *Space Shuttle Columbia* to avoid the density stratification caused by Earth's gravity.

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