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**MICROSCALE HYDRODYNAMICS NEAR MOVING CONTACT LINES**

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**ABSTRACT**

The hydrodynamics governing the fluid motions on a microscopic scale near moving contact lines are different from those governing motion far from the contact line. We explore these unique hydrodynamics by detailed measurement of the shape of a fluid meniscus very close to a moving contact line. The validity of present models of the hydrodynamics near moving contact lines as well as the dynamic wetting characteristics of a family of polymer liquids are discussed.

**INTRODUCTION**

Wetting and spreading of two immiscible liquids or a liquid and a gas across a solid surface are ubiquitous in nature and technology. In the reduced gravity of space, the behavior of multi-phase fluid systems can be greatly influenced, if not dominated, by forces associated with surface tension and the wetting characteristics of the system. The wetting of a surface is described by the shape of the fluid-fluid (liquid-liquid or liquid-gas) interface as it approaches the solid surface. The contact angle is often used to specify this shape near the solid surface and acts as a boundary condition to the differential equation describing the fluid-fluid interface shape. For static wetting, the correct determination of the contact angle hinges on the extrapolation of the hydrostatic shape of the macroscopic fluid body to the solid surface. The dynamic case is much more complex. Dynamic forces cause the stress field in the fluid and the curvature of the interface to increase as the contact line is approached.[1,2] This rapid increase of the interface curvature, called "viscous bending", makes it difficult to specify a contact angle in a manner similar to that used in statics.

To properly describe the moving contact line, one must alleviate the divergence of the stress field and interface curvature predicted when the hydrodynamics which successfully describes motion of a single fluid past a wall is applied up to the contact line. This is done by assuming that a small region near the contact line is governed by different hydrodynamics, such as allowing slip of the fluid past the solid surface. At present, we use a model consisting of an approximate expression for the shape of the fluid-vapor interface obtained by the method of

matched asymptotic expansions in three regions: (1) an inner region very near the contact line where the classical hydrodynamics break down, (2) an intermediate region further from the contact line which is controlled by the balance of viscous and surface tension forces and is governed by classical hydrodynamics, and (3) an outer region far from the contact line which is controlled by the balance of surface tension forces and gravity and is dominated by the macroscopic geometry of the fluid body.[3] The final description of the interface shape to order 1 in the capillary number  $Ca$  ( $Ca = \mu U / \gamma$  where  $\mu$  is the viscosity,  $U$  is the contact line speed and  $\gamma$  is the surface tension) is:

$$\theta \sim g^{-1}[g(\omega_0) + Ca \ln(\frac{r}{a})] + f_0(\frac{r}{a}, \frac{R_t}{a}, \omega_0) - \omega_0 \quad (1)$$

where

$$g(x) \equiv \int_0^x \frac{\rho - \sin \rho \cos \rho}{2 \sin \rho} d\rho ,$$

$r$  is the distance from the contact line,  $\theta$  the slope of interface,  $a$  is the capillary length,  $f_0$  is the outer interface shape for a static meniscus rising on a tube of radius  $R_t$  immersed vertically into a fluid bath in our experiments, and  $\omega_0$  is the single fitting parameter and characterizes the dynamic wetting of a materials system. The first term in equation 1, representing the interface shape in the intermediate region, and the value of  $\omega_0$  form the correct, dynamic contact angle boundary condition for the moving contact line problem.

## EXPERIMENTAL TECHNIQUE AND RESULTS

To experimentally probe the physics governing moving contact lines, we carefully measure the fluid-vapor interface shape very near a moving contact line.[4] Using videomicroscopy and digital image analysis, the local slope of the meniscus on the outside of a large diameter tube is determined from  $10\mu\text{m}$  to  $400\mu\text{m}$  from the contact line. Critical design and alignment of the optical system are essential to minimize systematic error in measuring the interface shape. Our system is calibrated by comparison to the known shape of static menisci.

To date, our studies have focused on the spreading of a class of polymer liquids, polydimethylsiloxanes (PDMS), on clean Pyrex.[4,5] We have found that equation 1 correctly describes the interface shape for  $Ca < 0.1$  (see figure 1) and that the predicted form of the interface shape in the intermediate region forms a geometry-independent boundary condition for the macroscopic interface shape in the outer region. Our results show that the asymptotic behavior of any model proposed to describe the unique hydrodynamics occurring in the inner region near the contact line must be of the form  $Ca \ln(r)$ . For  $Ca > 0.2$ , our measurements show systematic deviations from the model (see figure 2). These deviations are localized to a region near the contact line. The size of this region expands as  $Ca$  increases.

In a study of hydroxyl- and methyl-terminated PDMS, we have found that dynamic wetting is very sensitive to the details of the fluid-surface interaction.[5] Strong interactions of the fluid molecules and the surface enhance this sensitivity, and it is larger than the sensitivity to molecular weight. The  $Ca$  dependence of the parameter  $\omega_0$ , which describes dynamic wetting, is indistinguishable for methyl-terminated PDMS fluids which range over an order of magnitude in viscosity and are spreading over Pyrex surfaces with slightly varying surface chemistries (see figure 3). In contrast, the hydroxyl-terminated PDMS fluids which have a more chemically active end termination show distinct differences in their dynamic wetting depending on the precise chemical conditions on the Pyrex surface (see figure 4). These experiments show that the interaction between the polymer chain and the surface strongly affect the inner scale hydrodynamics.

Understanding the fundamental physics and chemistry governing moving contact lines and dynamic contact angles demands measuring properties free from the contamination of the specific macroscopic geometry of the experiment. In low gravity, the region where the geometry-free viscous forces dominate geometry-dependent forces expands. This region increases by over an order of magnitude from a fraction of the capillary length ( $a = 1.5\text{mm}$ ) on earth to a fraction of the experimental container (5cm) in low gravity. Thus, while optical systems detect only a small fraction of the geometry-free region in terrestrial gravity, they probe long extents of the region in low gravity. Thus, microgravity studies will be an important tool in future explorations of the geometry-free properties of moving contact lines.

#### REFERENCES

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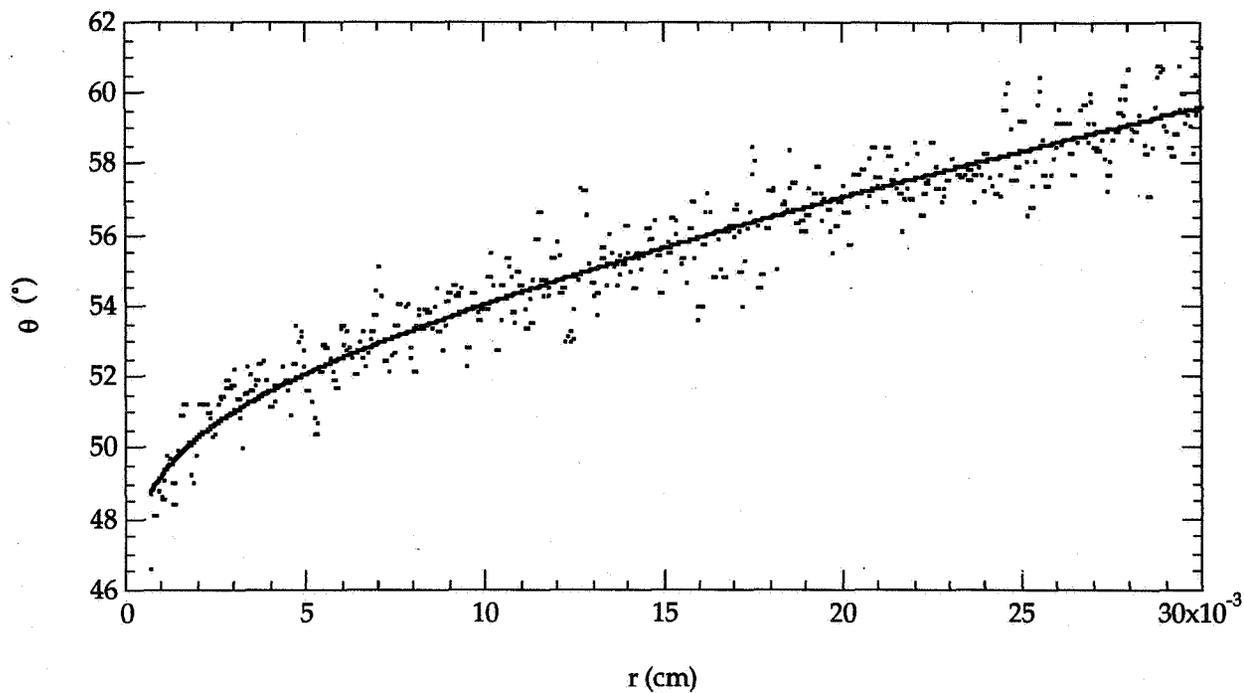


Figure 1. Interface shape data and fit at  $Ca = 0.005$ .

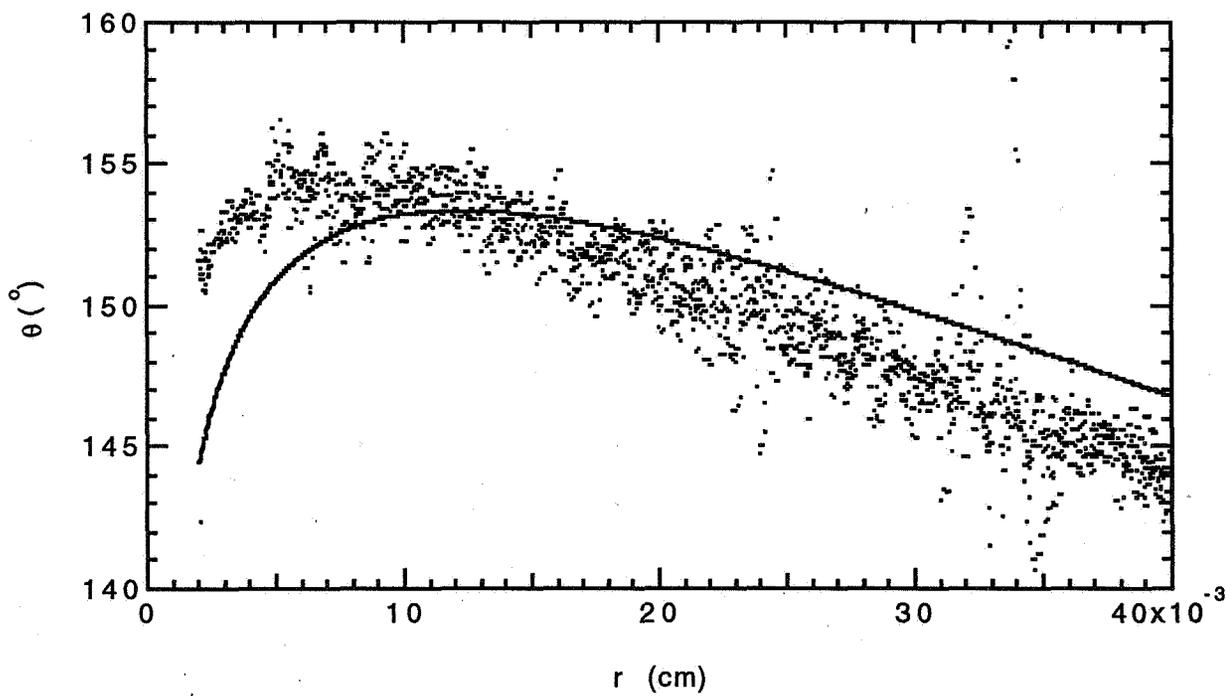


Figure 2. Interface shape data and fit at  $Ca = 0.46$ .

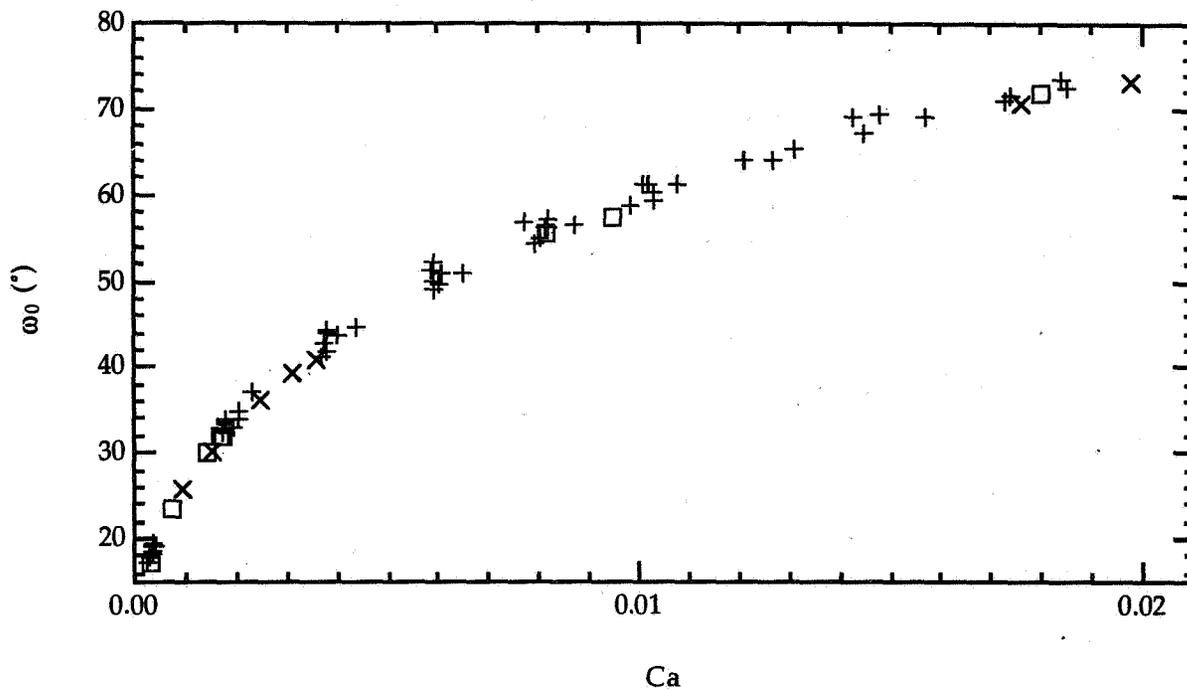


Figure 3.  $\omega_0$  vs.  $Ca$  for methyl-terminated PDMS of different viscosities on various Pyrex surfaces. (+ : 10poise, □ : 50poise, x : 100poise)

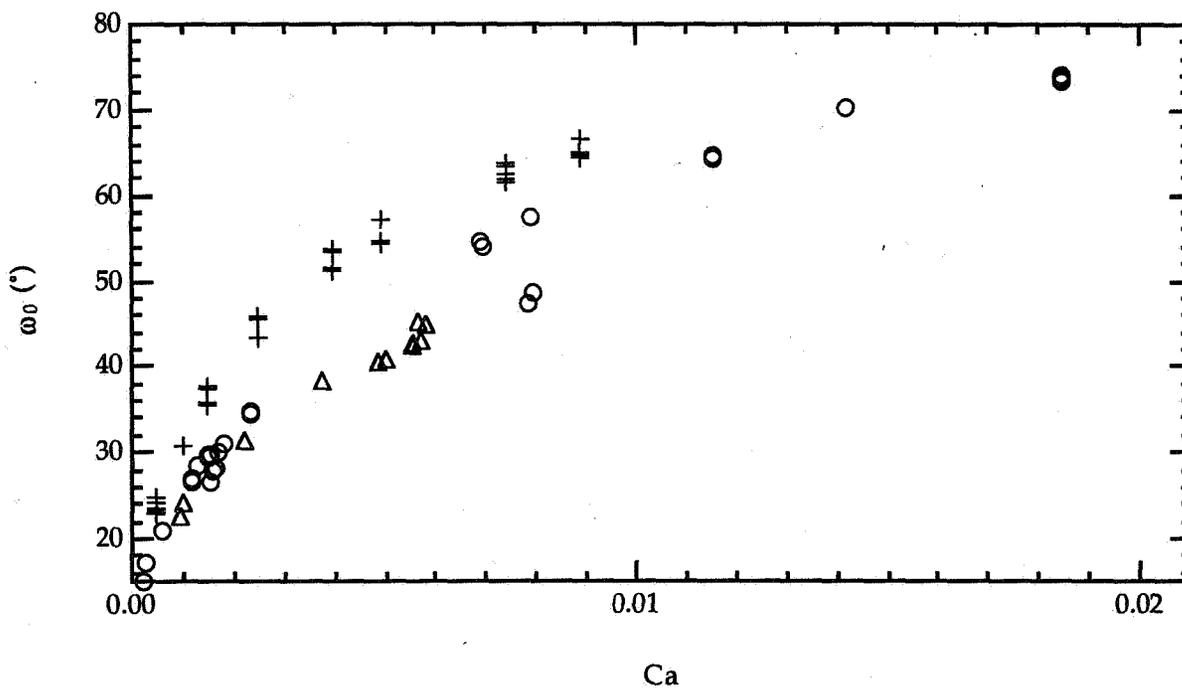


Figure 4.  $\omega_0$  vs.  $Ca$  for hydroxyl-terminated PDMS of different viscosities on various Pyrex surfaces. (+ : 10poise, O : 45poise, Δ : 160poise)