 FDMP, vol.4, no.1, pp.21-25, 2008

Precursor Film Length Ahead Droplet Traveling on Solid Substrate

I. Ueno[1], T. Konisho[2], T. Kawase[3] and T. Watanabe[4]

Abstract: The present authors carried out an experimental study with a special interest upon the dynamics of the fluid in the vicinity of the boundary line of three phases; solid-liquid-gas interface, which is so-called 'contact line.' The moving droplet on the solid substrate is accompanied with the movement of the boundary line of three phases; solid-liquid-gas interface, which is so-called macroscopic 'contact line.' Existing studies have indicated there is a thin liquid film known as 'precursor film' ahead the contact line of the droplet. In the present study the precursor film was detected by applying conventional ellipsometer, and its existing length was evaluated as a function of a dimensionless Capillary number. The present authors also indicate the effect of tiny particle sitting on the solid surface upon the traveling droplet and precursor film dynamics.

Keyword: Precursor film, wettability, droplet, Ellipsometry.

1 Introduction

Wetting and dewetting of the solid material by the liquid can be observed in phenomena in nature and technological applications; such as falling rain droplet on the window, boiling, thin film coating, crystal growth. These processes accompany with the movement of the boundary line of three phases, so-called 'contact line (CL).' Research on traveling droplet on a solid material has been widely carried out concerning the wettability problem; dynamic behavior and instability of the contact line in terms of static and dynamic contact angles (e.g., Bascom, Cottington & Singleterry (1964)), climbing/falling liquid film on inclined solid wall (e.g., Dussan V. & Chow (1983)), spreading of the droplet impinged on a solid material (e.g., Fukai, Shiiba, Yamamoto, Miyatake, Poulikakos, Megaridis & Zhao (1995)), etc. Through the experimental work, existence of thin precursor film ahead the contact line was revealed by interference microscopy and ellipsometry (Bascom, Cottington & Singleterry (1964)). Theoretical and numerical works employed an assumption that there be thin precursor liquid film of constant thickness ahead the CL. That is, the droplet travels on a quite thin liquid film, not on the solid surface. The dynamics of the fluid near the CL, however, has not been fully understood. Especially on the existing length of the precursor film, there exist few works by theoretical (Hervet & de Gennes (1984)) and experimental (Kavehpour, Ovryn &.McKinley (2003)) approaches. In the present paper the authors discuss the existing length of the precursor film by employing new approaches.

2 Experiments

Experimental apparatus is shown in Fig. 1. The relative variation of the height of liquid profile in the vicinity of the moving contact line was measured by use of conventional laser ellipsometer. A tiny liquid droplet of 5-cSt silicone oil was set on the inclined silicon wafer substrate, and the droplet traveled on it. The droplet was irradiated by elliptically polarized light of DPSS laser (λ = 532 nm).

Interferogram was observed by CCD camera I at the frame rate of 30 fps. The substrate on which the droplet sat was fixed at a constant incident angle of 45°. The whole interferometry system was

[1] Tokyo University of Science, Noda, Chiba, Japan
[2] Graduate School, Tokyo University of Science, Noda, Chiba, Japan
[3] Undergraduate, Tokyo University of Science, Noda, Chiba, Japan (present: Denso Corp., Aichi, Japan)
[4] Graduate School, Tokyo University of Science, Noda, Chiba, Japan (present: JTEKT Corp., Aichi, Japan)

placed on the L-shape rail, and the pitch angle of the rail itself, θ, could be varied; thus one can change the angle of the normal vector of the substrate to the horizontal surface without changing the laser incident angle. The motion of the liquid in the vicinity of the contact line was also observed with white light by another CCD camera (camera II) simultaneously to detect the position of the macroscopic contact line.

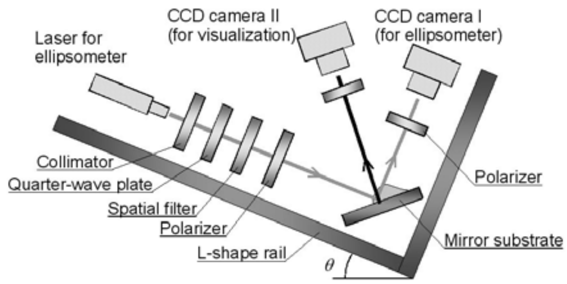

Figure 1: Experimental apparatus

Before each experimental run, several images of the fringe pattern without any droplet were obtained by CCD camera I. The image of fringe pattern for the evaluation was obtained by subtracting averaged background image without the droplet from the original recorded image with the droplet.

In the present study, the detection of the precursor film was conducted by applying two different ways; (1) to observe any kind motion of tiny particles sitting on the substrate ahead the traveling droplet, and (2) to observe the variation of the fringe pattern at a fixed point. All experiments were carried out under the room temperature condition (25 °C).

3 Results & Discussions

Wide-angle snapshots of the moving droplet are shown in Fig. 2; (a) the fringe pattern observed by camera I, and (b) the observed image with white light by the camera II. Each frame is digitally distorted in order to adjust the dimension. The droplet travels from the left to the right. The dashed line in the frame (a) indicates the 'macroscopic' contact line detected from the frame (b). One can clearly observe the fringe pattern in front of the macroscopic contact line as well, which indicates there must be a precursor liquid film with a non-uniform thickness. In a main series of experiments the region in the vicinity of the contact line was observed with a more zoomed field of view.

A typical example of the behavior of the particle sitting ahead the traveling droplet is shown in Fig. 3. This figure indicates a result in the case of water-silicon wafer system with frozen coffee particle. The particle sits on the top region in each frame, and the droplet travels upwards, which appears from the sixth frame. Each frame is captured at an constant interval of 2.0 s. One can clearly see the frozen coffee starts to melt before the macroscopic contact line of the water droplet reaches the particle. The authors also carried out a series of experiments in the silicone oil-silicon wafer system with several kinds of particles (diamond, silicon, etc.), and found that the particle exhibits a slight movement before the arrival of the macroscopic contact line.

It is noted here that the particle never moves or melt without putting the droplet on the substrate; the authors confirmed in preliminary experiments that the particle was not affected by the droplet located near the substrate (not on it). Any kind of change on the particle behaviour was observed only when the droplet is travelling towards the particle on the substrate.

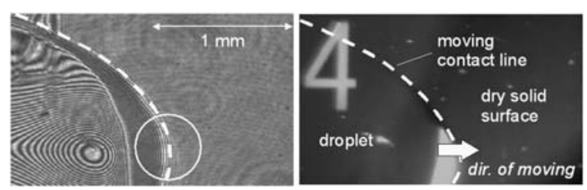

Figure 2: Snapshot of moving droplet in the vicinity of contact line in the case of silicone oil - Al mirror system; the fringe pattern (left: obetained by CCD camera I in Fig. 1) and the observed image with white light (right: by CCD camera II). Dashed line corresponds to the macroscopic contact line (M-CL). Fringe pattern lied in the right-hand region in the circle indicates an existence of 'precursor film' ahead the M-CL.

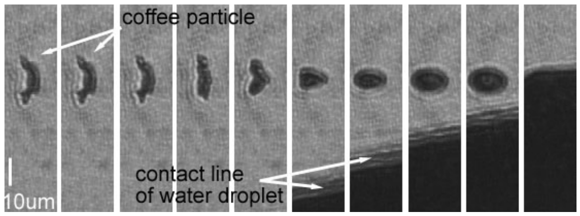

Figure 3: Typical example of a series of snapshots of fringe pattern obtained in the case of silicon-water system with frozen coffee particle. The droplet travels upward in this figure. The coffee droplet melts before the macroscopic contact line reaches it. The frame interval is 2.0 s.

Figure 4 shows a typical example of a snapshot of the spatial distributions of the interferogram (top) and its brightness data (bottom) in the direction of the normal vector of the macroscopic contact line. In the figure the droplet moves from left to right. The dashed line indicates the position of the macroscopic contact line. Through the reconstruction of the droplet profile from the fringe pattern, it is found that the bulk droplet travels on the substrate maintaining its shape almost the same; the advancing contact angle is almost constant. Ahead the macroscopic contact line, one can see fringe pattern that varies as a function of time. This indicates there exists a precursor film of non-uniform thickness.

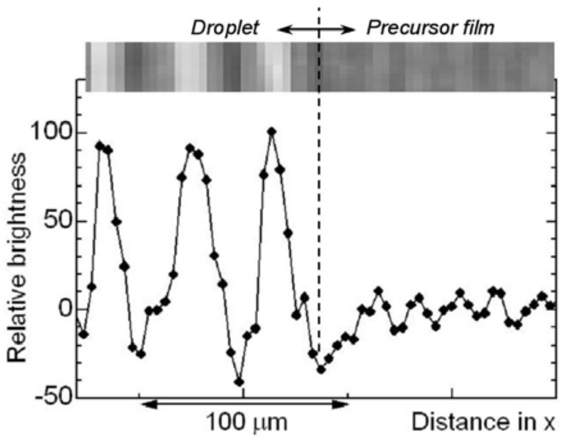

Figure 4: Brightness data in the vicinity of the macroscopic contact line (M-CL). Dashed line corresponds to the position of the M-CL.

In order to evaluate the existing length of the precursor film, a time series of brightness data from the fringe pattern at a fixed point is extracted. The present authors obtained the brightness by averaging four-pixel data. Figure 4 shows a typical example of the time series of brightness; the horizontal axis indicates a frame number corresponding to the elapsed time. The interval between the frames is 1/30 s. The brightness data is obtained before the droplet appears in the visualized field. The brightness data without any droplet on the substrate exhibit a spectrum of white noise. As the droplet comes closer to the measuring point, the brightness data fluctuate with a certain component as seen at the frames of 1500 $\sim$ 2300. As the macroscopic contact line of the droplet passes the measuring point, significant scatter of the brightness is observed at $\sim$ 2400 in the present figure. By checking the movie of the fringe pattern one can see a vague fluctuation as the droplet approaching. In the time series of the fringe pattern at a fixed point, however, it is rather difficult to determine the very initial point of the fluctuation due to the passing of the precursor film.

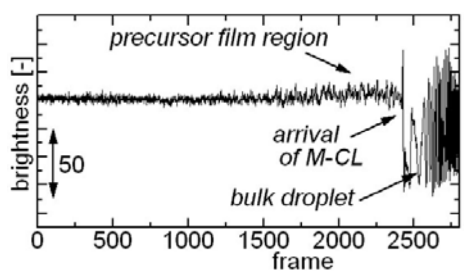

Figure 5: Example of time series of 4-pixel averaged brightness data on a certain fixed point before/after the droplet passes. The bulk droplet passes the measuring point at frame $\sim$ 2400.

A wavelet transformation with Morlet wavelet is applied to detect the initiation of the fluctuation. Figure 6 presents a typical example of the wavelet power spectrum after the transformation of the signal shown in Fig. 5. Huge power appears as the bulk droplet passes the measuring point at $\sim$ 2400 as aforementioned. Now one can notice that additional scale components of about 20 emerge at around the frame of 1300 (circled). This appearance of the additional components may correspond to the passing the front of the precursor film.

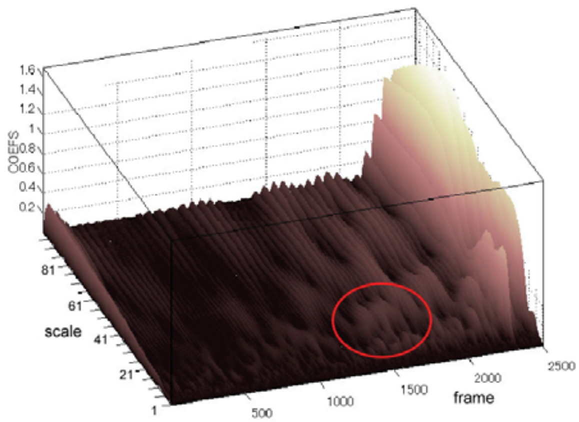

Figure 6: Wavelet power spectrum after transformation of the signal shown in Fig. 5 with the Morlet wavelet. The macroscopic contact line of the bulk droplet passes the measuring point at frame $\sim$ 2400. Additional scale components of about 20 emerge at around frame $\sim$ 1300 as indicated by the circle, which may arise due to the pass of the precursor film.

The distance between the macroscopic contact line and the detected region of the precursor film where the fringe pattern brightness starts to fluctuate is evaluated by detecting the position of the macroscopic contact line through the successive images obtained by the CCD camera II. The evaluated distance in the case of θ = 0 ° is plotted in Figure 7. In the figure the travelling speed of the macroscopic contact line is indicated as the non-dimensional Capillary number defined as Ca$\equiv \rho \nu U_{CL}/\sigma$ after the references, where ρ is the fluid density, ν the fluid viscosity, and σ the surface tension. It should be noted that the authors carried out a series of preliminary experiments to find out that the advancing speed of the macroscopic contact line, U_{CL}, is proportional to $t^{1/10}$, which follows the Tanner's law (Tanner (1979)). The solid and dashed lines indicate the theoretical prediction (Hervet & de Gennes (1984)) and the empirical equation (Kavehpour, Ovryn & McKinley (2003)), respectively. Although the present results indicate rather longer length comparing to the prediction and the preceding experiment, one can detect the length of growing precursor film of the same order as predicted value with the present way. In addition the present results do follow the prediction in which the length is proportional to Ca^{-1}.

Now the authors evaluate the precursor film length ahead the droplet sitting on an inclined substrate. Figure 8 indicates the evaluated length in the case of θ = 5 °.

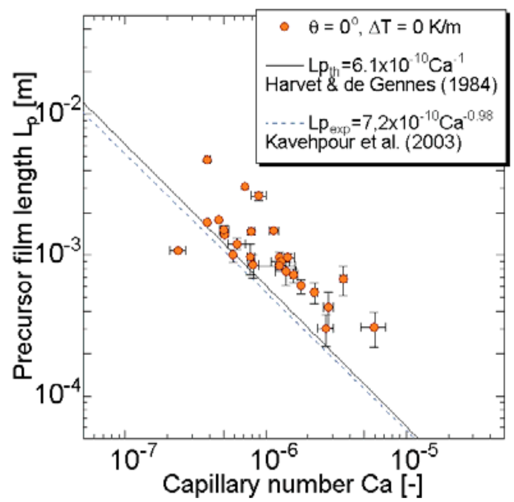

Figure 7: Precursor film length as a function of non- dimensional number Ca$\equiv \rho \nu U_{CL}/\sigma$, where ρ is the fluid density, ν the fluid viscosity, U_{CL} the traveling speed of the macroscopic contact line, and σ the surface tension. This figure indicates the results in the case of the droplet sitting on a horizontal surface. Solid and dashed lines indicate theoretical prediction (Hervet & de Gennes (1984)) and empirical equation (Kavehpour, Ovryn & McKinley (2003)), respectively.

One can clearly see that the detected length in the case of θ = 5 ° is longer than that in the case of θ = 0 °. Noted that the slope of the L_p against the Capillary number is almost proportional to Ca^{-1}, which agrees with the predicted value by the theoretical work as introduced in Figure 7. When the droplet is spreading on an inclined substrate, the macroscopic contact angle becomes an advancing one at the contact line moving downward. This variation results in a different curvature of the

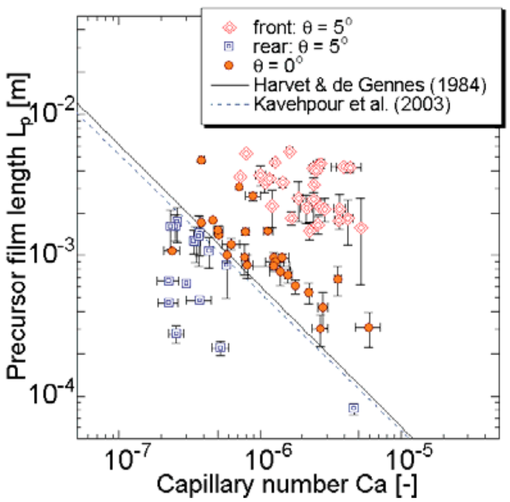

Figure 8: Precursor film length ahead the droplet sitting on a inclined surface as a function of non-dimensional number Ca$\equiv \rho \nu U_{CL}/\sigma$. The results in the case of horizontal surface, the preceding theoretical and empirical results as shown in Figure 7 are also plotted.

droplet surface from the one in the case of θ = 0 °, and in a larger amount of the fluid near the contact line. Thus, the fluid in the vicinity of the advancing macroscopic contact line is exposed to the variation of the capillary pressure and/or the variation of the static pressure compared to the case of the horizontal surface. It should be so hard to examine each effect independently on a terrestrial system. The authors group has been planning to carry out a parabolic flight experiment in order to check the effect of the gravity.

4 Concluding Remarks

The present authors carried out an experimental study on wetting process; especially focusing on the dynamics of the fluid in the vicinity of the macroscopic contact line of the traveling droplet on the solid substrate by employing conventional laser ellipsometry. The existing length of the precursor film ahead the macroscopic contact line was evaluated by applying wavelet transformation for the time series of the relative brightness data at a fixed point. The present experimental results indicate a longer length of the precursor film than the prediction by the theoretical work and the empirical value.

Acknowledgement: The authors acknowledge Dr. Atsushi Komiya (Tohoku Univ.), Dr. Satoshi Matsumoto (Japan Aerospace Exploration Agency (JAXA)) and JAXA for their invaluable cooperation for conducting the series of experiment. The authors gratefully acknowledge Dr. Komiya, Dr. Tatsuto Kimura (Kanagawa Univ.), Prof. Shigeo Maruyama and Dr. Junichiro Shiomi (The Univ. of Tokyo), and Dr. Matsumoto for fruitful discussions. Mr. Kanji Hirose (Tokyo Univ. of Science) is acknowledged for his devoted cooperation for carrying out a series of experiments. A part of the present study was financially supported by Grant-in-Aid by the Ministry of Education, Culture, Sports, Science and Technology.

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