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# Hysteresis Phenomena in a Liquid Sessile Drop on a Stretchable Inclined Elastic Substrate

V.A. Dekhtyar[1,A], A.E. Dubinov[2,A,B]

[A] Russian Federal Nuclear Center – All-Russia Research Institute of Experimental Physics, 37 Mira Avenue, Sarov, Nizhny Novgorod Region 607188, Russia
[B] Sarov Institute of Physics and Technology – Branch of National Research Nuclear University "Moscow Engineering Physics Institute", 6 Dukhova Street, Sarov, Nizhny Novgorod Region 607186, Russia

[1] ORCID: 0000-0001-8890-612X, valerik128@mail.ru
[2] ORCID: 0000-0002-3185-7926, dubinov-ae@yandex.ru

**Abstract**

The authors studied states of liquid sessile drops on a stretchable inclined elastic substrate to find out peculiarities of the contact angles hysteresis by digital optical microscopes. The drop elongation and contact angles were recorded on the stages of the band stretching and contraction. An anomalous behavior of the drop was found on the first step of the contraction during which the measured drop parameters remained unchanged. Finally, a new hydrophysical effect was found. It is hysteresis of the drop states, on the background of which the contact angle hysteresis exists.

**Keywords**: sessile drop, inclined elastic substrate, contact angle hysteresis.

## 1. Introduction

The contact angle hysteresis is a fundamental hydrophysical phenomenon which appears at the contact of liquid drops and a solid surface [1,2]. It reveals itself in the inequality the advancing contact angle of $\theta_A$ and the receding contact angle $\theta_R$ of the drop on this surface.

Hysteresis of contact angles can be recorded in different configurations [3,4]. The most technically simple configuration is the configuration in which the study of static and dynamic behavior of the drop is carried out on the inclined plane [5-7]. The study of hysteresis on the inclined plane can be carried out at the constant drop volume. Then, it is always true $\theta_A > \theta_R$ in the statics conditions for contact angles both in the case of wetting and nonwetting. However, sometimes the inversion of this inequality is possible at the drop sliding up on the inclined plane [8].

Recently, a new direction in the study of wettability and capillary phenomena has been developed. It is elastocapillarity in which the liquid behavior at the contact with elastic deformed surfaces is studied [9-11]. In the frames of this direction, the behavior of liquid sessile drops on the plane horizontal stretchable rubber surfaces was studied, for example, in [12,13]. Unique data about the hydrodynamics of drops was obtained in the research. However, it is clear that the hysteresis of contact angles in static conditions on the horizontal surfaces is absent. Thus, the hysteresis in [12] appeared evidently in a dynamic mode, for example, when the drop volume was changed by means of a syringe, as recommended in[2-4]. The hysteresis of contact angles in [13] was not studied at all.

Recently, the states of liquid drops sitting on a horizontal elastic cyclically stretched and then weakened substrate were studied experimentally [14]. A thin rubber band was used as a substrate, and glycerin was used as a liquid. A multi-branch hysteresis of the drop states was detected. The number of branches in the hysteresis can be adjusted by changing the time program for stretching-–loosening the substrate tension.

Thus, the aim of this work is to study a behavior of liquid sessile drops on a stretchable inclined elastic substrate to find out peculiarities of the contact angles hysteresis in the conditions when the drop volume is permanent.

## 2. Materials and methods

A rubber band was chosen as an elastic substrate for research (we used a famous rubber Martens' bandage; bands of necessary sizes were cut from it). Each band was of the following sizes: the length was 15 cm, the width was 1.5 cm, and the thickness was 0.5 mm. The band had a smooth work surface with less than 100 μm roughness. (Fig. 1).

A 46% water glycerol solution was chosen as a liquid, the producer was OJSC "Samaramedprom" (Russia) (hereinafter referred to as the "glycerin"). The glycerin was slightly colored to improve the visualization quality. It is known that glycerin is a weakly volatile liquid. Thus, the change of the drop volume during the experiment (less than 1 min) can be neglected.

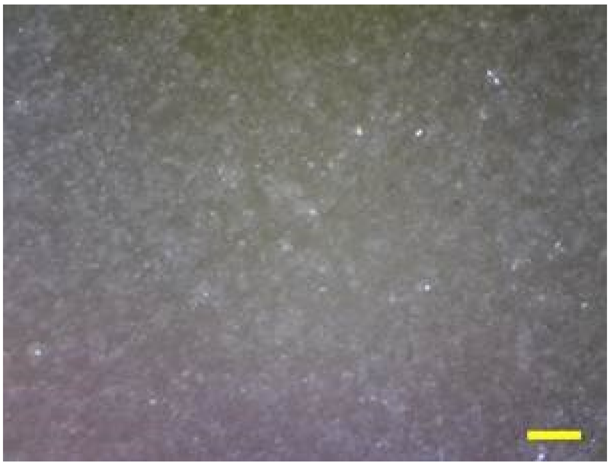


Fig. 1. Micro image of the work surface of a nonstretched rubber band (scale bar is 100 mm) obtained by means of the optical digital microscope 'Levenhuk' (MODEL D50L NG) [15].

Glycerin does not wet this rubber sample. The result of the contact angle measurement in the glycerin drop on the nonstretched horizontal rubber substrate is shown in Fig. 2. It is $99^o$.

The main experiments were carried out in the following way. A mark of a specified length was drawn on the band edge. It was possible to control the band elongation with the help of this mark. The band was placed on the plane inclined table before the experiment. One of the band ends was fixed, and then the band was stretched along the table surface until the elongation of $\delta \geq 2$. After that, a glycerin drop was placed on the work surface of the stretched band near the mark. Then, the force holding the band in the stretched condition was smoothly decreasing up to zero during 7–8 s (the $1^{st}$ step); some seconds left, the band again was stretched up to the elongation of $\delta \geq 2$ (the $2^{nd}$ step); and finally, the band was completely free from the stretching force (the $3^{rd}$ step).

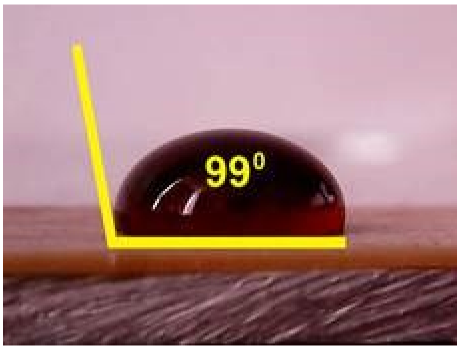



Fig. 2. The results of the contact angle measurement in the glycerin drop located on the horizontal rubber surface; the rubber band is not stretched (obtained by means of the optical digital microscope 'Celestron' (MODEL #44302-A) [15]).

The dynamics of the drop sliding deformation was recorded by means of the optical digital microscope 'Celestron' (MODEL #44302-A) [15] in the video recording mode with the frequency of 20 fps. The following values were measured on each frame: the mark size, the length of the drop base $l$, and the angles $\theta_A$ and $\theta_R$ (Fig. 3). The variables $l$, $\theta_A$, and $\theta_R$ characterize the drop state.

The velocity of the mark relative length change was not more than 0.1 $s^{-1}$ on each step (the 1st–3rd) in the experiments. Thus, the measurement of the drop geometry parameters $l$, $\theta_A$, and $\theta_R$ can be considered quasi-stationary; the Landau-Levich effects of the liquid drag by means of the moving substrate [16,17] can be neglected.

All the experiments were carried out at the room temperature of 18$^o$C and normal air pressure of 750 Torr.

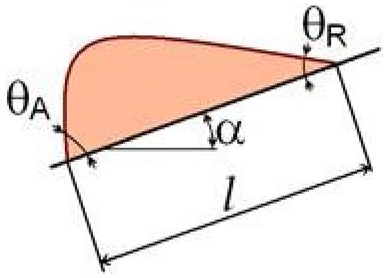



Fig. 3. The scheme of the drop located on the inclined plane; the measured values are explained here.

## 3. Experimental results

After the described experiment had been repeated several times, the following typical drop behavior was noticed. The form, the base sizes and the contact angles are changed slightly; the drop itself is as if sliding on the squeezed band (a sliding mode) during the 1st step. The drop elongates together with the band when the band is stretched on the 2nd step. When the band is again squeezed, the drop base is also decreased on the 3rd step. The drop edge remains as if stuck to the band surface along the whole perimeter on the 2nd and 3rd steps (a pinning mode).

The diagram of the dependence of the drop base relative length $\lambda = l / l_A$ and the band elongation d is presented in Fig. 4. Here, $l_A$ is an initial size of the drop base corresponding to the A point on the diagram. Fig. 4 illustrates the drop behavior. The diagram is constructed for one of the experiments; the table slope angle is 20$^o$, and the values of the diagram points are obtained after a scene selection of the video recording and frames analysis. The characteristic points B, C and D are also stated in the diagram; they are close to the ends of the 1st, 2nd, and 3rd steps, respectively.The roughnesses of curves on the diagram are connected with unremovable pixel errors of images.

There are two strait dotted lines on the diagram in Fig. 4: a horizontal line of the ideal sliding and an inclined line of the ideal (without sliding) drop drag by means of the moving band. It is evident on the $1^{st}$ step that nevertheless the drop is sliding on the band with a little drag, since the diagram is a little above the line of the ideal drag.A complete drop drag by means of the band does not happen on the $2^{nd}$ and $3^{rd}$ steps since the diagram is a little below the line of the ideal drag and there is a little sliding.

The drop states corresponding to the points A, B, C, and D are shown in video frames in Fig. 5. It is evident that the drop can be in different states (Fig. 5a,c) having one and the same value of the band elongation d; it depends on its prehistory. Thus, a new hydrophysical effect was found in this research. It is hysteresis of the drop states on the inclined elastic stretchable substrate.

We carried out several experiments in which the $2^{nd}$ and $3^{rd}$ steps of stretching and squeezing were repeated with one and the same drop many times. Finally, it was defined that the drop state passes along the diagram line between the C and D points in Fig. 4 many times, and the drop state passes between the A and B points only once, on the $1^{st}$ step.

We also carried out several experiments without the $1^{st}$ step, i.e. the drop was placed on the nonstretched band, and the hysteresis of states did not happen. It means that the drop motion on the $1^{st}$ step has a special character, at which the drop drag by means of the squeezed band is practically absent.

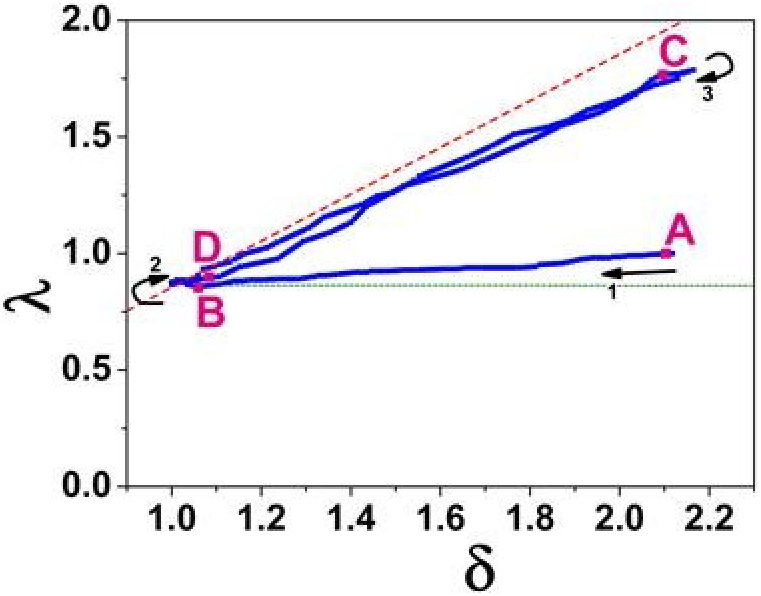
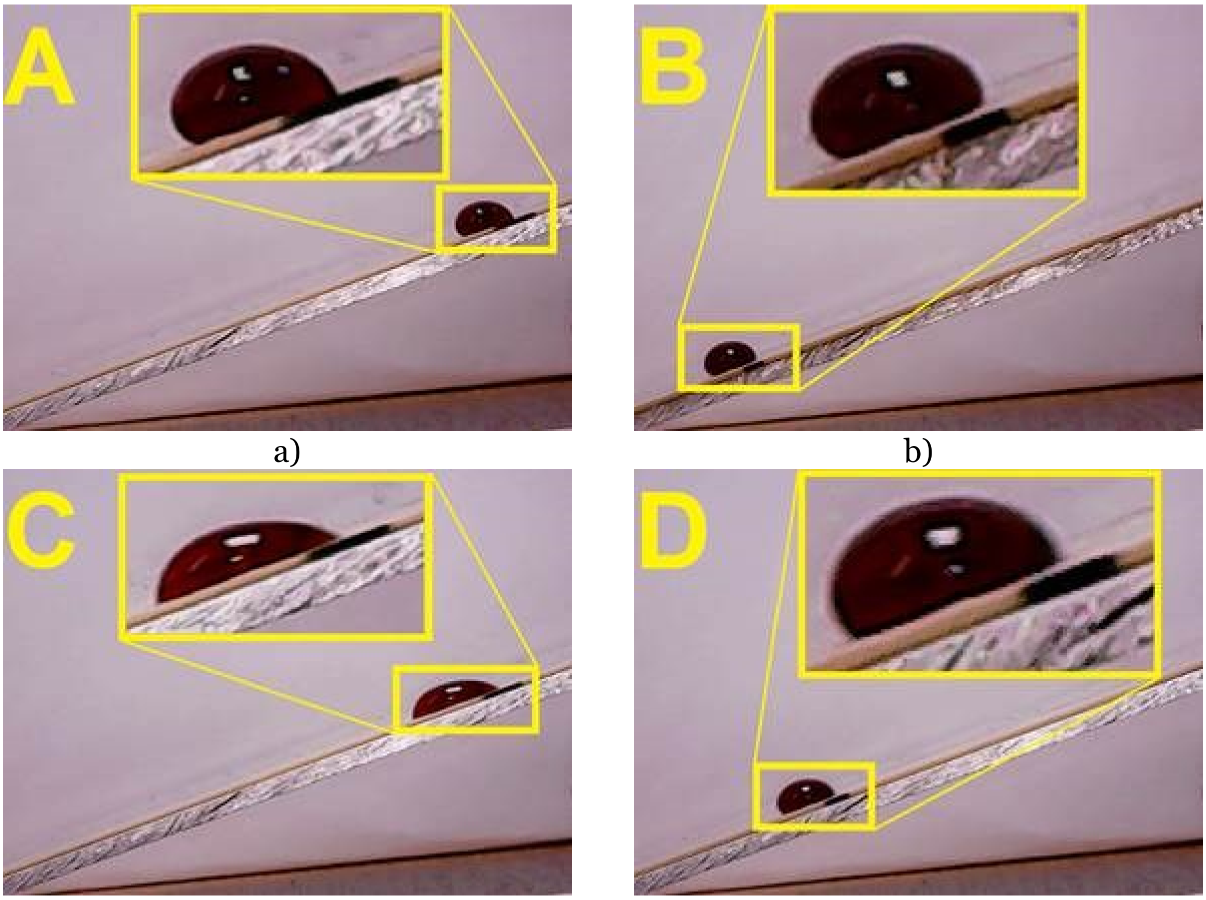



Fig. 4. The diagram of the dependence of the drop base relative length $\lambda = l / l_A$ and the band elongation d. Points A, B, C, and D correspond to the start of the $1^{st}$, $2^{nd}$, and $3^{rd}$ steps and some frames from the video recording in Fig. 5; the numbered arrows show the start of the corresponding steps; the horizontal dotted line is a line of the ideal sliding, the inclined dotted line is a line of the ideal drop drag (without sliding) by means of the moving band.

The diagrams of the dependence of the contact angles $\theta_A$ and $\theta_R$ and the band elongation d are presented in Fig. 6. They illustrate the hysteresis evolution of contact angles at the band deformation. It is evident that $\theta_A$ is below the conditional boundary dividing the areas of wettability ($\theta_A < 90^o$) and non-wettability ($\theta_A > 90^o$) at the initial moment, while $\theta_R$ is above this boundary. $\theta_A$ grows on the $1^{st}$ step and exceeds the level of $90^o$ at the end of the step, and $\theta_R$ is almost stable. Both contact angles $\theta_A$ and $\theta_R$ cross the level of $90^o$ on the next $2^{nd}$ and $3^{rd}$ steps. These crossings correspond to the transition from wettability to non-wettability and vice versa.

A special character of the 1st step is also evident here, it corresponds to the found hysteresis of states; besides, this hysteresis of states happens together with a famous hysteresis of contact angles.




a) b)

c) d)

Fig. 5. The drop states corresponding to the steps: a) – the initial state of the drop (point A in Fig. 4); b) – the drop state at the end of the 1st step(point B); c) – the drop state at the end of the 2nd step(point C), d) – the drop state at the end of the 3rd step (point D), the place and the length of the mark are evident under the drop base; the images are obtained by the optical digital microscope 'Celestron' (MODEL #44302-A) [14].

## 4. Conclusions

The authors studied the states of liquid sessile drops on a stretchable inclined elastic substrate to find out peculiarities of the contact angles hysteresis.

The authors carried out the experiments in which the drops volume remained stable in every experiment. A special weakly volatile liquid – glycerin was used for this purpose. A rubber band was used as a substrate.

The band was placed on the plane inclined table before every experiment. One of the band ends was fixed, and then the band was stretched along the table surface up to the specified elongation. A glycerin drop was placed on the work surface of the stretched band. After that, the force holding the band in the stretched condition was smoothly decreasing up to zero (the 1st step); then some seconds left, the band was stretched again (the 2nd step), and finally, the band was completely free from the stretching force (the 3rd step).

The drop elongation and the contact angles were recorded. An anomalous behavior of the drop was found on the 1st step during which the measured drop parameters remained unchanged. Finally, a new hydrophysical effect was found. It is hysteresis of the drop states which happens together with a famous hysteresis of contact angles.

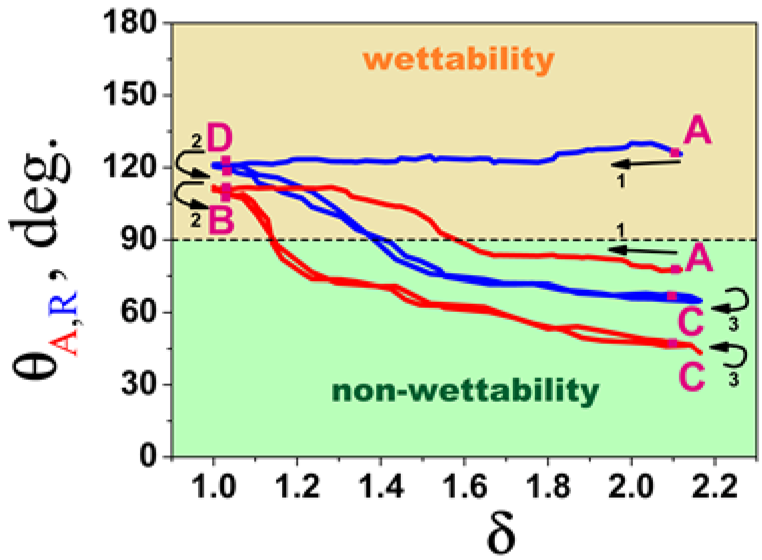



Fig. 6. The diagrams of dependence of the contact angles $\theta_A$ and $\theta_R$ and the band elongation d. Points A, B, C, and D correspond to the start of the 1st, 2nd, and 3rd steps and some frames from the video recording in Fig. 5; the numbered arrows show the start of the corresponding steps; the horizontal dotted line is a conditional boundary dividing the areas of wettability ($\theta_{A,R} < 90^o$)andnon-wettability ($\theta_{A,R} > 90^o$) .

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