Notes

Design and Testing of a Low-Cost and Compact Brewster Angle Microscope

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Received September 6, 1995

Introduction

Since its introduction by two groups in 1991[1,2] the Brewster angle microscope (BAM) has come into widespread use for the study of monolayers, both on liquid and on solid surfaces. This is not surprising, as the BAM can reveal inhomogeneities, particularly phase domains in ultrathin layers, without the use of special probes. This is a clear advantage over other techniques, such as for instance fluorescence microscopy, which require the addition of fluorescent probe molecules that may disturb the local environment of the probe and thereby cause artifacts.[3] Since more and more monolayer studies on systems of increasing complexity are being carried out, a BAM should be a compact, easy-to-operate instrument. Instruments that are presently commercially available have good performance but are rather heavy and relatively expensive.[4] We have therefore developed a BAM that is as compact as possible, with high optical quality. In addition, its construction from standard optical parts can be easily realized at moderate cost.

Design Objectives

The operation of the BAM is based on the well-known fact that, for a pure Fresnel interface, the reflectivity of the in-plane polarization component becomes zero at the Brewster angle. Hence, when a surface of, e.g., pure water, which is an almost pure Fresnel interface, is illuminated under the Brester angle with monochromatic p-polarized light, the reflectivity can be as low as 10^{-7}; i.e., it will appear dark. The reflectivity is not exactly zero because of some surface roughness caused by capillary waves. The presence of very small amounts of material on the water surface will affect the Brewster condition so that (in most cases) the reflectivity goes up. Regions differing in density or orientation of molecules on the surface will therefore show up by virtue of their optical contrast.

One problem inherent to the BAM is that the surface must be viewed under an angle. In a conventional linear arrangement of the viewing optics, it is then impossible to focus simultaneously on the entire area of interest. Only a narrow strip of this area will be in focus; the farther away surface elements are from this in-focus strip, the more fuzzy they will appear. Moreover, viewing under an angle obviously yields images compressed along the line of sight. Meunier[2] solved these problems by confocal scanning of the area of interest, retaining only narrow-strips around the focal line. The final image was then composed of these strips after they had been treated digitally to remove the compression. This leads to good images, but the disadvantage is that taking and processing the entire image is a time consuming operation, which requires that imaged objects do not move during capture. The only way to suppress the collective motion of the molecules at the surface is to use a very small trough which is protected from disturbing air currents by a cover. We would prefer an instrument that can be used with standard commercial Langmuir troughs, where such precautions cannot be easily taken.

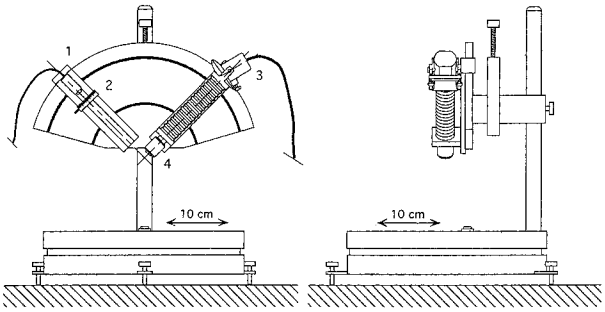

Figure 1. Brewster angle microscope setup. Numbers refer to the following parts: (1) diode laser including collimator; (2) dichroic sheet polarizer; (3) CCD camera without lens; (4) microprojection objective.

Another technical difficulty is that reflectivities, even in the presence of molecules on the surface, are very low, typically of order 10^{-5}. In order to cope with this, one can of course increase the illumination intensity and/or try to enhance the detection sensitivity. Meunier used a sensitive CCD camera in combination with an Ar^+ ion laser, but the drawback of this is a rather bulky instrument.

Last, but not least, care should be taken to avoid parasitic background contributions from scattered and reflected light; these spoil the contrast of the image. In most cases, this problem can be solved by blocking or absorbing the ray that is refracted into the subphase.

In our design, we wanted to achieve good contrast and sensitivity without using heavy or bulky parts. Also, we wanted to avoid the kind of image (re)construction used by Meunier. Below we describe our setup.

Description of the Setup

Our BAM consists of a small diode laser (LaserMax MDL-200-680-35, 34 mm long, 11 mm diameter) which emits 35 mW at 680 nm. The emitted beam passes through a polarizer (Melles Griot 03 FPG 001) and then illuminates a spot of about 2 mm^2 on the surface. The reflected light passes an objective (which can be chosen according to the desired range of magnification) and is finally detected by a CCD camera (COHU model 6710). Both the objective lens and the CCD camera are somewhat tilted with respect to the line of sight, which partly removes the compression of the final image, while keeping it in focus (Scheinpflug arrangement). It is this combination of a Scheinpflug arrangement and a fairly long focal distance that allows

(1) Hönig, D.; Möbius, D. *J. Phys. Chem.* **1991**, *95*, 4590−4592.
(2) Hénon, S.; Meunier, J. *Rev. Sci. Instrum.* **1991**, *62*, 936.
(3) Lösche, M.; Sackmann, E.; Möhwald, H. *Ber. Bunsen-Ges. Phys. Chem.* **1983**, *87*, 848.
(4) Hönig, D.; Möbius, D. *Thin Solid Films* **1992**, *210/211*, 64.

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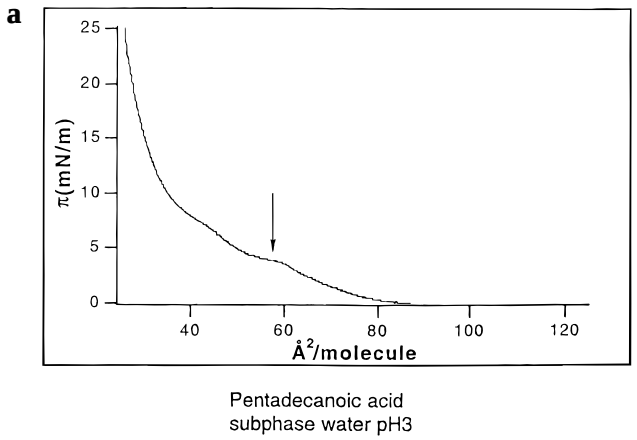

Figure 2. Surface pressure isotherm (a) and BAM images (b and c) of pentadecanoid acid (PDA) on water at pH 3. The images were both taken at the surface density indicated by the arrow in part a. Part b was taken immediately after compression; part c was taken after some extra compression followed by expansion. Image size: 610 × 920 μm^2.

us to take the entire image at once without too much distortion. The camera has its maximal response at 680 nm and has a lower detection limit of 0.0125 lux. Its output has 512 horizontal TV lines and 415 vertical TV lines. The observed area at maximum magnification is 500 × 360 μm^2, so that 1 pixel of the camera corresponds to about 1 μm^2. The resolution is typically a few micrometers. Forstoring the data, we have both a framegrabber (Data Translation DT55-EZ-50), which typically captures about two images per second, and a video recorder, which operates at 25 frames/s.

Both the light source and the imaging optics are mounted on arms which can be pivoted around the reflection point, so that the angle of incidence can be adjusted to the Brewster angle of the substrate used. The entire microscope is mounted on an X-Y sliding table. With

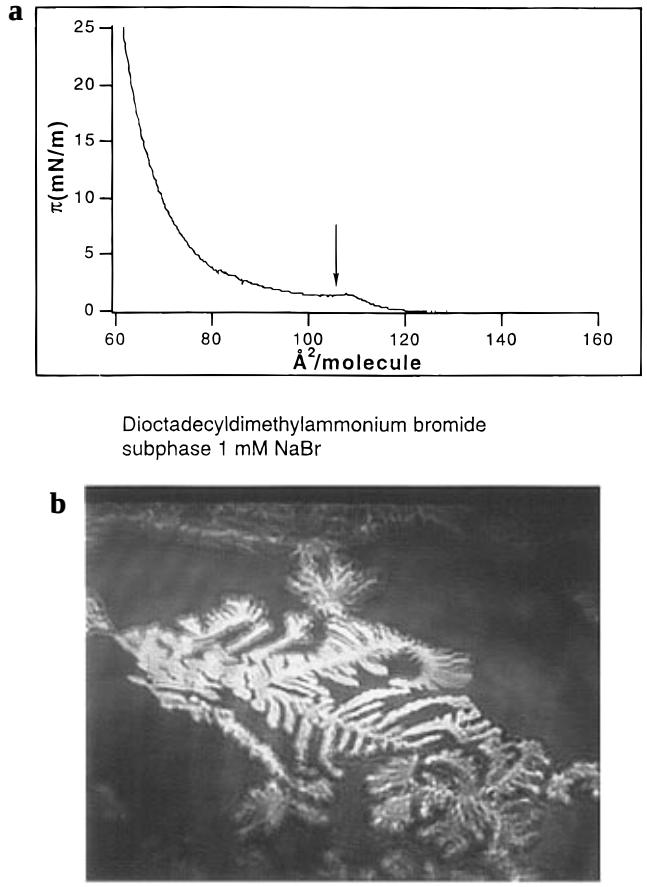

Figure 3. Surface pressure isotherm (a) and BAM image (b) of dioctadecyldimethylammonium bromide (DODMA) on a 1 mM aqueous NaBr solution. The image was taken at the surface density indicated by the arrow in part a. Image size: 610 × 920 μm^2.

the help of this it can be positioned over a usual Langmuir trough, and the surface can be viewed during a (de)-compression. It is best to use a symmetrical trough with two moving barriers, because in this case the microscope can be placed in the center of the trough and the surface moves only little during (de)compression. The setup is shown in Figure 1.

Testing

Two different monolayer forming amphiphiles were used for testing the setup: pentadecanoic acid (PDA) on water of pH 3 as the subphase and dioctadecyldimethylammonium bromide (DODMA) on 1 mM NaBr. They were taken on a standard open Langmuir trough with a total area of 1100 cm^2, equipped with two moving barriers. The microscope was positioned in the center between the two barriers. We show the π/A isotherms of both systems (Figures 2a and 3a, respectively), two images of PDA (Figures 2b and c), and one image of DODMA (Figure 3b), taken in the two-phase range of the isotherm at a point indicated by the arrow. All images have a size of about 700 × 550 μm^2.

The first picture taken from the PDA monoalyer (Figure 2b) shows the familiar circular patches that the condensed phase of this molecule forms.[4,5] This shape is expected for liquid-like phase domains with a sufficiently strong line tension. The size of the patches varies here between 40 and 250 μm (cross section), and from the length of the plateau in the surface pressure isotherm (from 60 to about 50 Å^2/molecule) we estimate that the density of the

(5) Hönig, D.; Overbeck, G. A.; Möbius, D. *Adv. Mater.* **1992**, *4*, 419.

molecules within the patches is approximately 20% higher than that in the surrounding dilute (LE) monolayer. Since the intensity of the reflected p-wave is to first order linear in the molecular density, the average contrast in the picture should be of the same order. It can be seen that such a difference is easily resolved. Note that the patches do not have a uniform intensity but do have domains slightly different in intensity. These are probably domains within the patches that differ in molecular orientation. Our second image (Figure 2c) was taken after some expansion. Now the LE phase nucleates within the patches, forming circular dark holes.

For the condensed phase of DODMA we observe a much more ramified structure, as can be seen in Figure 3b; this agrees with observations reported by others.[6] Dendritic structures like these form when the dense phase is solid-like; they may originate from a diffusion-limited growth mechanism. Again, the contrast is fairly good, and most of the image is reasonably in focus. Details with sizes of about 10 μm are readily viewed. Note that we take images here at the acquisition time of the video camera, so that kinetic studies should be quite feasible.

Conclusions

We have constructed a simple easy-to-operate Brewster angle microscope at very reasonable cost that allows us to study the phase behavior of monolayers on standard commercial Langmuir troughs. Although there is room for further improvement, the tests with well-studied systems show that the setup as presented here is certainly capable of following the formation of all kinds of surface phases just as fast as a video camera can operate.

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(6) Ahuja, R. C.; Caruso, P.-L.; Möbius, D. *Thin Solid Films* **1994**, *242*, 195.