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Extending the surface force apparatus capabilities by using white light interferometry in reflection

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(Received 2 June 2003; accepted 21 August 2003)

An important factor in the success of the surface force apparatus (SFA) in measuring interactions between surfaces over nanometer separations has been the optical interference technique used to measure the surface separation. Until recently, this technique has only been used when both of the materials are transparent. As a result, thin sheets of mica have been the material of choice. We describe a simple method to extend the capabilities of the SFA so that a wide variety of material surfaces can be studied while retaining an optical measurement technique. The key to this technique is to modify the optics so that reflected, rather than transmitted, light is used to produce the interference pattern. Now, only one material is required to be thin and transparent while the other can be any material providing it is at least partially reflective. To succeed with this technique, it is necessary to maximize the visibility of the interference fringes. This is achieved by optimizing the thickness of a partially reflective coating (often silver) deposited on the back side of the transparent material. © 2003 American Institute of Physics. [DOI: 10.1063/1.1619551]

I. INTRODUCTION

The physical forces that act between surfaces of materials in close proximity are vitally important in many products and processes in daily use. For the past quarter of a century, the surface force apparatus (SFA) has had outstanding success in characterizing surface forces, and in using them as a probe of solid surfaces and the behavior of thin fluid films. It was developed initially by Tabor’s group to study van der Waals forces in air and a vacuum, and later refined by Israelachvili and Adams to extend its usefulness to include interactions in liquids. The basic instrument has changed very little since its inception with its key components consisting of a force measuring spring, crossed cylindrical surfaces, mechanisms for moving the surfaces with a precision of 0.1 nm, and an optical interference method for measuring the separation between the surfaces. It is the last feature that this article concentrates on. The method for measuring separation in the SFA is optical interferometry that produces fringes of equal chromatic order (FECO). By using this method, the surface separation can be determined with a resolution of about 0.2 nm in the traditional SFA arrangement of crossed cylindrical mica surfaces.

FECO are produced when collimated white light is shone normally into a gap between two reflecting surfaces. The gap in the SFA usually consists of two mica sheets separated by some intervening medium. A thin layer of silver is deposited onto the back sides of the mica sheets to provide suitably reflective surfaces. As the gap thickness varies, different wavelengths within the spectrum interfere constructively. By directing the light transmitted through the gap to a spectrometer, the subsequent interference pattern can be viewed.

The advantage of using FECO is twofold. First, the absolute distance between the interacting surfaces can be measured accurately, because the contact (zero) position can be clearly identified. Second, the shape of the interacting surfaces is easily determined as it is directly related to the shape of the FECO pattern. The latter feature is of major benefit when studying adhesion and deformation as well as providing a useful method for determining the interaction geometry and identifying particulate contamination.

The use of FECO is also considered by many to be the main disadvantage of the SFA because it limits the materials available for study in the SFA to those that are transparent. The basis of this criticism seems to stem from the misconception that FECO in the SFA are only observed when light is transmitted through the surfaces. Consequently, most studies performed with the SFA have concentrated on using pairs of thin (1–3 μm) transparent sheets of mica. In an effort to explore materials other than mica in the SFA, a number of modifications to the instrument have been devised. These include: Coating the mica with a thin layer of another material; preparing other transparent materials to replace the mica; and substituting the optical thickness measurement with a nonoptical technique. The last approach has resulted in hybrid instruments, like the MASIF and those developed by Tonck et al. and Salmeron et al., that have capabilities that lie between the SFA and the other important force measurement alternative, the atomic force microscope (AFM).

Although the aforementioned techniques go some way to alleviating the limitations imposed by transparency requirements, a more common solution to this problem is to use a different instrument to measure forces between opaque surfaces. Derjaguin et al. used a force balance to measure the...
force between crossed fibers while others study the colloidal interaction directly by exploiting techniques, such as total internal reflection microscopy and video microscopy. However, the most popular approach is to use the AFM, particularly since the introduction of the colloid probe technique by Ducker et al. Like the SFA, the AFM monitors the deflection of a cantilever to determine the interaction force.

Unlike the SFA, the contact position in an AFM measurement has to be inferred from the spring deflection data using the region of constant compliance to define a zero position. This problem is not unique to the AFM but is common to many of the techniques using nonoptical methods to determine the surface separation, including those implemented in modified SFAs. Even though these instruments often provide very precise measurements of the relative surface separation, not knowing the contact position can present difficulties in interpreting results. When contamination or adsorbed layers are present, these may be misinterpreted as contact, and interpreting results. When contamination or adsorbed layers are present, these may be misinterpreted as contact, and when the surface is deformable, a region of constant compliance is not easily identifiable. The other related problem is that of determining the geometry of the interaction in situ. Avoidance of these two problems is an important advantage of the SFA. Hence, the apparent success of these techniques in overcoming the problem of probing other types of surfaces must be tempered by the loss of information for accurately determining the contact position. In the traditional SFA, this is provided by the optical technique.

Here, we offer another approach to addressing the problem of studying opaque surfaces in the SFA that retains the benefits of the optical distance measurement. All that is required is a modification of the optical arrangement so that rather than observing FECO in transmitted light, reflection fringes are used. In doing so, the lower surface can be almost any material providing it is partially reflective and sufficiently smooth. Although the upper surface must still be transparent, existing techniques can be exploited to provide a range of surfaces capable of probing an even greater range of materials at the lower surface. Using this method, resolutions between 0.2 nm and 5 nm have been achieved depending on the reflectivity of the lower surface. We also note that Spikes has used reflection FECO successfully to monitor lubricating films confined between a rolling ball and flat surface down to thicknesses of 1–2 nm.

In this article, we describe how the SFA can be modified to incorporate reflection FECO and discuss the requirements that must be considered in order to analyze the data. Examples of FECO produced in reflection are provided to demonstrate the quality of the fringes that have been observed and the distance resolution that can be obtained.

II. METHOD

The essence of our approach is to modify the optics so that rather than observing transmission FECO, we use reflection FECO. However, there are a number of associated issues that must be considered when doing this. First, the quality of the fringes depends on the reflectivity of the surfaces. Poorly reflecting surfaces produce fringes that have low contrast and low finesse (they are relatively broader than for their highly reflecting counterparts). Second, more often than not, the arrangement of the different layers within the interferometer will be asymmetric, in which case analytic methods for converting the fringe wavelength to separation are not available. These difficulties are tackled using digital image processing and analysis techniques to extract fringe positions from fringe patterns of variable quality. The issue of converting wavelengths to separation is dealt with using numerical modeling of the interferometer. It is this combination of reflection optics, image processing, and numerical modeling of the interferometer that ensures the technique is useful.

A. Fringes of equal chromatic order in reflection

The principles of the formation of reflection FECO are essentially the same as for transmission FECO. There are however a few differences in the nature of the fringes. The most obvious of these is that reflection FECO appear as dark bands on a bright background. This is a consequence of observing the results of destructive interference (reflection FECO) rather than constructive interference (transmission FECO). The other important, though subtle, difference has to do with the path taken by light entering the interferometer relative to where it is observed. Figure 1 shows a comparison of the path the light takes when interacting with the interferometer for transmission and reflection fringes, respectively, as well as the associated intensity pattern. Light transmitted through the interferometer is a combination of a ray that passes completely through the interferometer and subsequent rays that have undergone multiple reflections before exiting the interferometer. The reflected light, on the other hand, consists of a ray that is reflected from the top surface and never enters the interferometer, in addition to the subsequent rays that have undergone multiple reflections before exiting through the top of the interferometer. The first reflection can have an important effect on the minimum intensity and, hence, the contrast of reflection fringes. To optimize the contrast of the fringes, it is necessary to select a silver thickness that is thick enough to be reflective and enhance the interferogram of the interferometer and the respective intensity profiles (below) for two cases: (a) When both surfaces are semitransparent and light is transmitted through the surfaces and (b) when one of the surfaces (e.g., Hg) is opaque and light is reflected off this surface. In the transmission case, the dashed line indicates the first reflection from the surface of the lower mirror, which is never collected. However, in the reflection case, although we are only interested in the light reflected from within the interferometer, it is not possible to avoid collecting light from the first reflection. Another important point to note is that for some materials, and in particular metals, some of the light will be absorbed, which results in phase changes at these interfaces.
For each of the systems, there is an optimal silver layer thickness for this interferometer, as well as the caveat is that the quality of the results relies on the accuracy of the chosen optical constants. The lower graph is a plot of intensity normalized by the incident intensity as a function of silver layer thickness. In all cases, the following interferometer parameters were kept constant: \( t_{\text{mica}} = 4000 \text{ nm}, t_{\text{aqueous}} = 10 \text{ nm}, t_{\text{Hg}} = 500 \text{ nm}, \) and \( t_{\text{decane}} = 5 \times 10^2 \text{ nm}. \) Literature values are used for the refractive index of silver (Ref. 29), the mica \( \beta \) line (Ref. 30), mercury (Ref. 31), and decane (Ref. 32).

FIG. 2. Example calculations using the MMM demonstrating the effect of silver layer thickness on fringe visibility (contrast). The upper graph shows intensity profiles calculated for a \( \text{Ag|mica|aqueous|Hg} \) system with different silver layer thicknesses: \( t_{\text{Ag}} = 0 \text{ nm} \) (chain line), 20 nm (solid line), and 40 nm (dashed line). Some points to note are the shift in minima position due to phase shifts (Refs. 27 and 28) and the sharpening of the fringe that accompanies an increasing silver layer thickness. The lower graph is a plot of contrast (defined here as the difference between maximum and minimum intensity normalized by the incident intensity) as a function of silver layer thickness for three different systems: \( \text{Ag|mica|aqueous|Hg} \) (squares), \( \text{Ag|mica|aqueous|air} \) (circles), and \( \text{Ag|mica|aqueous|decane} \) (triangles). In all cases, secondary fringes produced from light reflected within the layer are produced which tends to make them easier to identify. Another important challenge that poorly reflecting surfaces present is the associated problem of an increased presence of secondary fringes produced from light reflected within the top material only.\(^{27}\) The latter arrangement results in broad low contrast fringes and consequently makes locating and analyzing the fringes difficult. To help alleviate this problem, thicker mica (6 to 8 \( \mu \text{m} \)) is used so that the narrower fringes are produced which tends to make them easier to identify. Another important challenge that poorly reflecting surfaces present is the associated problem of an increased presence of secondary fringes produced from light reflected within the top material only.\(^{27}\)

While the calculation illustrated in Fig. 2 gives a guide to the ideal thickness, experimental trial and error is recommended for further optimization of fringe quality. In general, when the lower surface has a moderate to high reflectivity (e.g., a silicon wafer or mercury drop), sharp fringes can be obtained using silver films that are around 20 to 30 nm thick. In the case of poorly reflecting surfaces (e.g., the air/water interface), much thinner silver layers are usually required (~7 to 10 nm). The latter arrangement results in broad low contrast fringes and consequently makes locating and analyzing the fringes difficult. To help alleviate this problem, thicker mica (6 to 8 \( \mu \text{m} \)) is used so that the narrower fringes are produced which tends to make them easier to identify. Another important challenge that poorly reflecting surfaces present is the associated problem of an increased presence of secondary fringes produced from light reflected within the top material only.\(^{27}\)

As with transmission FECO, valuable information about the shape of the interacting surfaces can also be extracted from reflection FECO. Figure 3 illustrates how this is achieved. This aspect of FECO interferometry is a key advantage of it over other interference techniques utilizing monochromatic light as well as noninterferometric measurement methods.
B. Experimental arrangement

In our system, a cold white light source is provided by a 150 W globe coupled to a fiber optic bundle (FOT 150 Fiber Optic P+P AG Spreitenbach, Switzerland). A simple lens at the tip of the bundle helps to roughly collimate the light, after which the light is directed through a 50/50 beam splitter to the surfaces. Before it reaches the surfaces, the light passes through a microscope objective. This performs the job of focusing the reflected light from the interferometer into the entrance slit (~80 µm wide) of a scanning spectrometer (Thermo Jarrell Ash Model 82050 0.5 m Ebert Scanning Spectrometer, with a 590 groove/mm grating Genesis Laboratory Systems, Inc., Grand Junction, CO) via the beam splitter. A polarizer, placed between the beam splitter and spectrometer, is used to remove one of the birefringent lines when mica is used as the transparent material. The spectrometer is recorded from the output of the spectrometer using an intensified Vidicon camera (VE-1000 SIT, Dage-MTI, Michigan City, IN). This has a horizontal resolution of 750 lines and operates at video frame rates (PAL: 25 frames/s or 50 frames/s interlaced). The camera signal is digitized using a video frame grabber (Fidelity 200 DT3852-2, Data Translation Inc., Marlboro, MA) to produce an 8-bit grayscale image of 756 × 568 pixels (giving a minimum effective wavelength resolution of 53 pm/pixel). Nonlinearities in the camera output are carefully calibrated and corrected before spatial and wavelength data are extracted from an image. Wavelength is calibrated using the green and yellow spectral lines of mercury. Equilibrium measurements involve recording a set of images directly to a personal computer and analyzing these postexperiment while dynamic measurements are recorded on video tape using a Sony UVW-1400P Betacam SP (Sony Corp., Tokyo, Japan) with individual frames grabbed and analyzed off line.

C. Fringe analysis using image processing

From the earlier discussion on the importance of the silver layer, it is apparent that the quality of fringes largely depends on the reflectivity of the surface being studied. In many cases, the fringes may not be as sharp as often observed for silvered mica sheets. Rather than measure the fringe position manually, which becomes increasingly difficult as the fringes get broader, image processing techniques are used to extract this information. There are also the additional advantages of consistency between measurements, the ability to measure on multiple fringes, and the ease of collecting large amounts of data which is particularly useful for obtaining detailed shape information or analyzing dynamic effects.

An effective method of locating the fringe position automatically, which forms the basis for a number of image processing algorithms described in the literature,33–36 is to fit an appropriate curve to the extrema in the measured intensity profile. In our system, we adopted the algorithm of Quon et al.35 mainly because it is designed to also extract the shape of the fringe (which is particularly advantageous when studying deformable fluid surfaces).37 Because we are interested in minima rather than maxima in the intensity profile, the data are inverted to suit the software. Having done this, the image is binary thresholded to isolate the parts of the intensity profiles corresponding to fringes from the background followed by fitting a Gaussian curve to each profile to extract the extrema corresponding to fringe positions.

When grabbing the images for analysis (as opposed to recording illustrative examples of the fringes), some preconditioning of the camera signal is normally required. Where possible, signal averaging, either over several frames or by controlling the exposure time, is used to reduce background noise. We also adjust the analog to digital converter input parameters of the frame grabber to match the available digitization levels to the maximum amplitude of the camera signal containing fringe intensity information. This maximizes the signal-to-noise ratio and makes it easier for the image processing algorithm to detect the fringe positions.

D. Converting wavelengths to separation

Now, with the possibility of using a wide variety of materials within the SFA, the layers of the interferometer will often be arranged asymmetrically. In addition to this, one of the surfaces will potentially also provide the reflective layer. Therefore, analytic methods for calculating the separation will only be available in rare instances. A more general approach is to use the multilayer matrix method. Unfortunately, the MMM cannot be used to determine the layer thickness directly since it produces wavelength-dependent reflectivity, whose minima provide fringe wavelengths as a function of layer thickness, whereas the experiment yields a set of wavelengths that need to be converted to film thickness. The approach we adopted for solving this problem involves a two-step procedure. In the first step, the thickness of the transparent surface(s) is determined when the surfaces are in contact. Once this thickness is known a look-up table of separation as a function of wavelength is constructed as the intervening film thickness is varied in the MMM model from zero (contact) to the maximum anticipated thickness. By comparing the measured wavelengths with those in the look-up table, the experimental surface separation can be found.38,39

An advantage of employing the MMM is that it can be easily adapted into a tool for exploring the fringe patterns in unknown material combinations, as illustrated by Fig. 2. A further refinement involves using calculated intensity profiles to simulate images of fringe patterns for different geometries. This is achieved by building up the spectrometer from a set of intensity profiles calculated at different radial positions along the interferometer as indicated in Fig. 3. Heuberger et al.27 demonstrated the utility of this technique for studying secondary fringes in the standard SFA whereas here we use it to guide the experiment when searching for fringes in unfamiliar systems.

III. ILLUSTRATIVE RESULTS AND DISCUSSION

So far, the technique of using FECO in reflection has been used successfully to study interactions with mercury surfaces.37,38,40,41 We have also demonstrated that surfaces of other materials, including a silicon wafer, an air bubble,
and an oil drop, all separated from mica (or silica in the silicon wafer case) by an aqueous layer, can be studied using FECO in reflection. Figure 4 shows some typical fringes observed for these materials. With the exception of the fringes produced with mercury, the other interferometers have not been optimized and, with further development, the fringe quality and distance resolution may be improved. The point we want to make here is that it is possible to observe the fringes of interest (the dark curved lines) without too much difficulty.

Figure 5 shows an example of how reflection FECO can be used to study poorly reflecting deformable surfaces. In this case, the mica is rapidly driven toward an air bubble in water. As the two come closer, the hydrodynamic pressure between them causes the bubble to deform and adopt a dimple shape at its center. This process can be followed directly from the fringe shape. Using image analysis and the MMM, the fringe positions were converted to separations to produce the data shown. Importantly, the absolute film thickness, a factor crucial to correctly interpreting hydrodynamic data, is measured unequivocally. Unlike nonoptical measurements, this is obtained directly using the optical measurement. Clearly, with a combination of knowing the contact position accurately, reasonable resolution in the relative position, and a high density of data in the shape, we can produce data that provide detailed information about this process. This measurement is also an example of a worst-case scenario where only a limited amount of preprocessing is possible since each curve must be measured from a single video frame. In this situation, the resolution suffers because of the inherent noise in the grabbed image.

We estimated the surface separation measurement resolutions of the interferometers shown in Fig. 4 by calculating the root-mean-square (rms) standard deviation from a curve fit through parts of the extracted profiles. Because the Ag/mica/aqueous/Hg interferometer has been optimized, we obtain the best resolution of the four examples although the Ag/pyrex glass/aqueous/silicon system is comparable. When image averaging has been used, the resolution can be as good as 0.3 nm. In situations where it was not possible to employ image averaging, the resolution is about 0.7 nm for Ag/mica/aqueous/Hg, with no comparable data available for the system involving silicon. The next example involves using Ag/mica/aqueous/air and has a resolution of 1 nm (with image averaging) and 1.3 nm without averaging. Lastly, we have the Ag/mica/aqueous/decane interferometer which has a resolution ranging from 3–6 nm depending on whether video frames are averaged. Although significantly worse than the previous three cases, this also represents an extreme example of a poorly reflecting surface.

ACKNOWLEDGMENTS

The authors would like to extend their appreciation to Doug Smith, Ben Francis, Darren Bachmann, and Danny Edwards for developments in the MMM software. This work was supported by the Australian Research Council.
