The x-ray surface forces apparatus for simultaneous x-ray diffraction and direct normal and lateral force measurements

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(Received 9 October 2001; accepted for publication 1 April 2002)

We describe the experimental setup and principles of operation of the second-generation x-ray surface forces apparatus that allows for the first time simultaneous x-ray scattering and direct force measurements. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1480461]

Experimental techniques that can measure forces and simultaneously probe structure and structural evolution in confined thin films are promising assets for understanding interfacial processes. While force measurements and theoretical simulations suggested the occurrence of structure-related transitions as, e.g., oscillatory forces and stick-slip friction, these have not been *directly* proved experimentally using an independent probe. This is mainly due to the technical difficulties in performing spectroscopy, diffraction, or high-resolution microscopy of molecules that are confined and sheared between two solid surfaces. Recently, Kuhl *et al.* introduced a confinement cell for investigating complex fluids using neutron diffraction.[1] There has also been considerable effort in coupling friction experiments with a variety of *in situ* probes.[2] Particularly promising is the coupling of two well-established techniques; the surface forces apparatus (SFA) and x-ray diffraction (XRD). The first generation of the x-ray surface forces apparatus (XSFA), adapted directly from the SFA-III which was introduced by Israelachvili and McGuiggan,[3] allowed for the first time x-ray structural measurements to be carried out on confined complex fluid samples using a synchrotron source.[4] In the initial experiments, the XSFA was successfully used to study effects of shear and confinement in liquid crystalline films.[5–7] These experiments demonstrated the unique advantage of combining the SFA methodology with *in situ* x-ray small angle scattering, which provides direct information on molecular arrangements in the confined sample. However, the first generation XSFA did not permit any direct normal or lateral force measurements to be made simultaneously with XRD experiments. We have recently designed and constructed a experimental setup that allows, for the first time, for simultaneous direct lateral and normal force measurements and XRD. The capabilities of the upgraded XSFA were demonstrated in monitoring shear-induced orientational transitions in lyotropic liquid crystals.[8] In this Note, we present the experimental setup and principles of operation of the second-generation x-ray surface forces apparatus (XSFA-II).

A close-up photograph of the experimental setup of the XSFA-II on the in-house small angle x-ray scattering spectrometer (two-circle diffractometer) is shown in Fig. 1. The x-ray beam path is from right to left and the white light path from the fiber optic source in the front, to the optical stage in the back of the image. Also seen on the optical table behind the XRD are the grating spectrometer and long working distance objective lens mentioned above, and the portable halogen white light source (Chiu Technical Corp., Kings Park, NY, model FO-150).

A more detailed schematic is presented in Fig. 2, showing the XSFA-II chamber with the functional windows and ports which were designed for the simultaneous XRD and force measurement functions. A Be exit window was installed in the back of the chamber in order to minimize the attenuation of the weak diffracted beams. An ultrathin (30 μm) silica entrance window (Rayotek Scientific, San Diego CA) was mounted in the front of the chamber, to allow for visible and x-ray radiation to enter simultaneously. A sapphire optical window was positioned on the right-hand side of the chamber to allow for the white light to exit to the long

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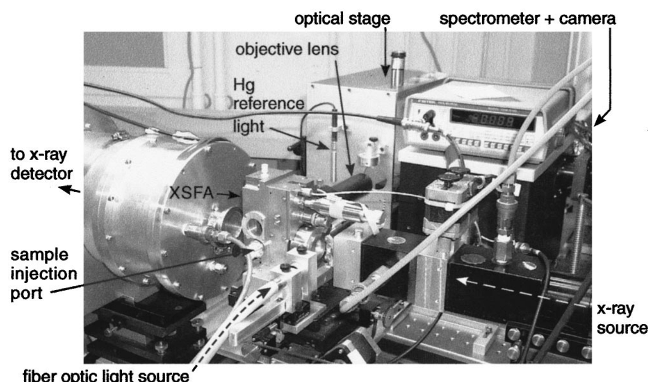

FIG. 1. The experimental setup of the XSFA-II, showing the path of the x-ray beam from right to left and the white light path from the fiber optic source in the front, to the optical stage in the back of the image.

working distance objective. A side port was used to inject the sample in between the two molecularly smooth mica surfaces, with an adjacent window for viewing the surfaces during and after sample injection. The XSFA-II chamber was designed for measurements in a well-controlled temperature, humidity, and chemical environment. For this purpose, the chamber was tightly sealed, and equipped with a thermocouple port, a liquid reservoir which can be filled with aqueous salt solutions to allow for a controlled humidity, or with organic solvents to achieve a controlled vapor environment. Entrance and exit valves allowed to purge the chamber with a flow of inert or reactive gas. The modified SFA chamber is mounted in the beam path of an x-ray diffractometer, while the rest of the white light interferometry components are setup on a separate, portable optical table. These included a long working distance objective (VZM 450, Edmund Scientific), custom designed optical stage, 0.34 m grating spectrometer (Spex Industries, Inc., model 340S) which was equipped with a viewing port and digital micrometer for electronically transmitting the fringe position, and a VE-1000-SIT silicon intensified tube low-light camera (Dage-MTI Michigan City, IN). The setup was designed so that both x rays and white light are directed in the same optical

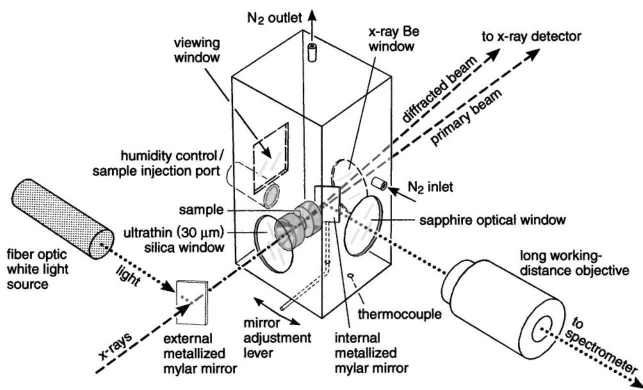

FIG. 2. Detailed schematic illustration of the XSFA-II, which allows all the basic capabilities of the SFA (including motion and force measurement in the lateral direction) simultaneously with x-ray diffraction. Note the two 45° Al-coated mylar mirrors used for reflecting the white light through the sample. While being transparent to x rays, these mirrors allow the use of x rays at the same time that white light interferometry is used.

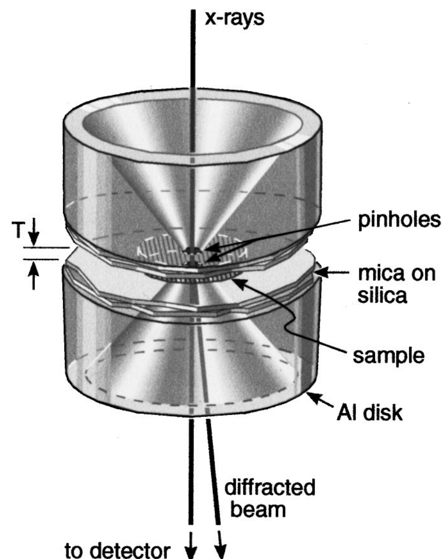

FIG. 3. Custom-designed cone-shaped aluminum disks for the XSFA-II, which are mounted in a crossed-cylinder geometry. The center pinhole allows for transmission of x rays through the sample. The back-silvered mica sheets are mounted on thin, curved silica sheets that provide solid support across the pinhole opening.

path, allowing for both XRD and multiple beam interferometry to be carried out in transmission. For this purpose, two adjustable, 30-μm-thick mylar mirrors coated with a thin layer of Al (Metallized Products, Inc., Winchester, MA) are positioned in the beam path at an angle of 45° to allow the steering of the light with a relatively small x-ray attenuation. The front mirror is mounted externally, between the front x-ray exit slit and the SFA chamber, while the back mirror is located inside the SFA chamber, immediately behind the back disk holder. Force measurement data is taken using a laptop personal computer, and interferometry fringes are viewed on a video monitor and taped on VCR. All components are portable, to allow the setup of the XSFA-II both at synchrotron and in-house x-ray radiation sources.

An important feature of the experimental setup is the custom-designed, inverted cone-shaped aluminum disk pairs which supported the thin mica sheets used in the XSFA-II. The outer surfaces of each of the disks had a cylindrical curvature of 20 mm, which when cross-mounted facing each other, effectively constituted a sphere-on-a-flat geometry (as in conventional SFA). In order to allow for transmission of x rays through the sample, the disks were equipped with concentric $\phi = 1.0$ mm pinholes which were mounted against each other on the x-ray beam path, as shown in Fig. 3. The back of the mica sheets is coated with a 55-nm-thick layer of Ag, and then mounted Ag side-down on thin, curved silica sheets that provide solid support across the pinhole opening. The silica sheets were glued to the aluminum disks using Norland optical adhesive 61 (Norland Products Inc., New Brunswick, NJ) which was cured for 10 min in a UVOX ultraviolet radiation source. The mica sheets were glued down onto the silica using EPON 1004 glue (Shell Chemical Corp.).

In order to carry out the lateral motion and force measurement capabilities of the SFA, a piezoelectric bimorph

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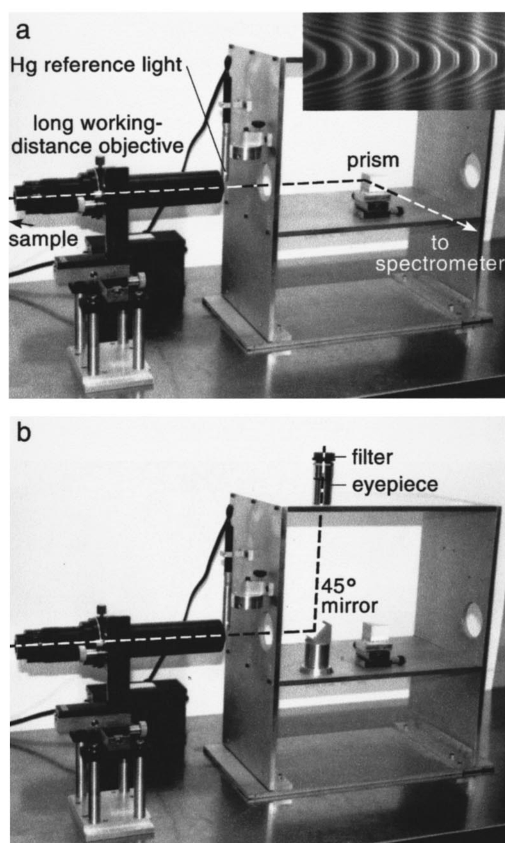

FIG. 4. The portable optical stage is used in two geometries: (a) interferometry mode. Light is guided from the objective lens to the spectrometer for analysis of fringes of equal chromatic order (inset). The Hg pen-light reference source on the left-hand side of the stage is used for determining the absolute fringe wavelengths. (b) Microscopy mode. A second mirror is inserted in the light path for guiding the light from the objective lens to a viewing eyepiece, which is located on a glass window on top of the optical stage.

slider[9] was used for laterally moving the front surface with displacement amplitudes of ~30 μm. Alternatively, a reversible dc motor driving a spring loaded antibacklash translation stage was used for laterally moving the back surface with displacement amplitudes of ~500 μm. A custom-made friction sensing device[10] was installed for measurement of lateral forces (not shown). This allowed to couple XRD with both rheological and tribological measurements when the surface separation is of the order or greater than the contact area (rheology) or much smaller than the contact area (tribology).

The multiple beam interferometer is the heart of the SFA optical system which allows for accurate gap distance measurements and normal force profiling in the SFA. For this purpose, a portable optical stage was designed for guiding the light from the XSFA chamber to the spectrometer. Figure 4 shows the two modes of operation of the optical stage. In interferometry mode, the light is guided from the long working distance objective to the spectrometer using a right-angle prism [Fig. 4(a)]. This allows to view fringes of equal chromatic order (FECO) and monitor changes in their wavelength. The inset in Fig. 4(a) shows a set of FECO obtained with the optical setup of the XSFA-II. Additionally, light from an Hg pen-light source is used for determining the *absolute* fringe wavelengths. Figure 4(b) shows the use of the optical stage in microscopy mode. In this mode, the combination of the long working distance objective lens with an ocular lens (viewing eyepiece) constitutes a simple microscope which is used for viewing the pinhole area in transmission. This proved critical for aligning the single asperity contact between the two mica surfaces within the pinhole area, i.e., in the area sampled by the x-ray beam. By adding a second 45° mirror after the objective lens, the light is directed to the eyepiece which is located on a glass window on top of the optical stage, as shown in Fig. 4(b).

In conclusion, we have presented the experimental setup of the XSFA-II for coupling direct force measurements with XRD, and demonstrated its use in monitoring shear-induced orientational transitions in lyotropic liquid crystals.[8] The design of the XSFA-II allows the use of both x-ray and visible light optics on the same optical path, with minimal compromise in x-ray attenuation and in the wavelength resolution of the multiple beam interferometer due to the portable optics used. While the current setup is designed for transmission small angle x-ray scattering, it is, in principle, possible to rotate the XSFA chamber and perform XRD in parallel with the mica surfaces (glancing incidence geometry). Furthermore, work is underway to improve the spatial resolution of the x-ray probe by using microfocusing x-ray optics for separately probing distinct sections in the contact with micron resolution.[11] The use of a third-generation synchrotron source will improve the time resolution of the XRD experiments, and allow combined structural and direct force measurements of films confined in much smaller gaps.

The authors are grateful to Dottie McLaren for expert artwork assistance. J.I. and Y.G. acknowledge financial support of the U.S.–Israel Binational Science Foundation Grant No. 9800395. This work is supported by ONR No. N00014-00-1-0214 and NSF No. DMR-0076357. The Materials Research Laboratory at UC Santa Barbara is supported by NSF No. DMR-0080034.

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