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Block Copolymer Films I

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Liquids under Shear Explored by Neutron Scattering: A Problem in Lubrication

Max Wolff, Rainer Hock and Andreas Magerl
Lehrstuhl für Kristallographie und Strukturphysik, Universität Erlangen-Nürnberg, Bismarckstr. 10, 91054 Erlangen, Germany.
Bernhard Frick
Institut Max von Laue-Paul Langevin, Avenue des Martyrs, 38042 Grenoble, France.
Hartmut Zabel
Lehrstuhl für Festkörperphysik, Ruhr-Universität-Bochum, Universitätsstr. 150, 44780 Bochum, Germany.

ABSTRACT

Considering the atomic scattering cross sections for neutrons they are an excellent tool to investigate lubrication problems. Two different shear cells have been built to investigate both the dynamics and structural properties of liquids under shear: one cell has been optimised for quasielastic and inelastic neutron scattering while another one has been designed for reflectivity and diffraction work. The dynamical aspects have been studied on the high-resolution backscattering instrument (IN16 at Institut Laue-Langevin (ILL)). Data with a commercial motor oil as a sample have been taken in contact with an aluminium boundary showing directly the developing anisotropy of diffusion under shear. Furthermore within the same set-up it has been possible to monitor the macroscopic velocity distribution including surface slip. In addition, a diffraction experiment has been carried out, demonstrating from a measurement of the position and the profile of the graphite 002 reflection that the ordering of macroscopic graphite particles in a flowing liquid can be studied with neutrons and an ordering with a tilt angle of the particles of 5° to the flow has been determined.

INTRODUCTION

It is estimated that about 6 % of the gross national product of the USA are lost due to friction and wear [1]. In spite of this fact the detailed mechanism of lubrication is not well understood up to now. Considering the high penetration power of neutrons for many engineering materials and the large scattering cross section of hydrogen contained in most lubricants neutrons appear to be an excellent tool to investigate lubricants in massive environments like cryostats, furnaces or shear devices under conditions relevant for applications. Arguing along this line it has been shown that the macroscopic flow of lubricants can be observed by neutron backscattering [2, 3, 4]. The investigations have revealed different velocity distributions between a fixed and a rotating disc depending on the adhesion to the surface. Such differences in the macroscopic flow should relate with structural changes as well as with changes in the microscopic diffusion processes, especially at the solid liquid interface. This contact region has recently attracted much attention. Differences from the normally assumed non slip boundary condition of liquids have been found by light scattering [5] and atomic force microscopy [6]. An unusual ordering of macroscopic particles near the surface under flow has also been reported by x-ray scattering [7]. In the present work we investigated the macroscopic velocity distribution of a commercial motor oil between a fixed and a rotating disk for a temperature of 18°C via neutron backscattering measurement,

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indicating strong surface slip. Within the same measurement we also gather information about a change from an isotropic diffusive motion of the liquid without shear to anisotropic diffusion in the sheared sample. To obtain a more complete picture we have also started diffraction work and show by the 002 Bragg reflection that macroscopic graphite particles suspended in paraffin order with a tilt angle of about 5° with respect to the flow direction.

EXPERIEMNTAL DETAILS

Neutron backscattering. High resolution neutron backscattering (IN16 at ILL) makes it possible to acquire information about the macroscopic flow of a liquid and obtain within the same measurement information about diffusion processes within the sample. The high-energy resolution of less than 1μeV is achieved by analyser crystals (Si 111) set to backscattering geometry. To make different energies the neutrons velocities are modified by a Doppler drive located before the sample. The speed of this Doppler drive is a few m/s, quite similar to the speed that is obtained with the shear apparatus.

For the investigation of liquids under shear, two different scattering geometries have to be considered. In reflection geometry the vector of momentum transfer is parallel to the surface normal and therefore perpendicular to the flow direction. Thus only information about the microscopic diffusion is obtained from quasielastic scattering. Turning into transmission geometry with the scattering vector parallel to the flow, in addition, the macroscopic motion of the liquid becomes visible from inelastic Doppler shifted neutrons. A special plate-plate shear device has been developed for these dynamic neutron experiments [4].

Neutron diffraction. At the instrument (ADAM at ILL) the graphite 002 Bragg reflection at the scattering vector Q=1.9 $Å^{-1}$ was investigated for two different suspensions of graphite in paraffin (20 ml and 75 ml mixed in 100 ml paraffin). A new kind of shear device has been constructed for neutron diffraction of liquids under flow. It has a single crystalline silicon plate and a rotating aluminium plate underneath, the gap of 0.5 mm being filled with the liquid [4]. This device makes it possible to investigate samples with the scattering vector Q in the direction of the surface normal, i. e. Q is orthogonal to the direction in the former diffraction work where Q is in the plane of the solid-liquid interface.

RESULTS

Neutron backscattering. Figure 1 presents high resolution back backscattering measurements. The first column (a-d) shows data for the empty cell with the aluminium discs made visible by putting a strong elastic scatterer like a tape on to them. This allows to determine the instrumental resolution to 1.2 μeV, which is slightly increased from the standard value of 0.8 μeV. The reason is a displacement of the detector from the exact backscattering position due to the large size of the shear device. The three pictures a, b, c on the left show data with both discs fixed in transmission and reflection geometry and with one disk set into rotation in reflection geometry. As expected there is no difference visible implying that only elastic scattering from the discs is visible. However, in transmission geometry and with one disc rotating, an additional inelastic peak from Doppler scattered neutrons appears at –7 μeV corresponding to a disc speed of about 1.5 m/s. This second peak is broader than the elastic one due to the velocity gradient from the finite beam size (in the present case a beam size of about 2.5 cm was used with a disc radius of 9 cm). If needed the beam area can be optimised in size to reach the maximum resolution provided by the instrument. However this will result in a loss of neutron flux and prolong the time necessary to obtain one dataset (the individual spectra shown took about 24 h each). The different intensities of the two peaks arise from different amounts of tape glued on the stationary and the rotating disc and has no further physical meaning.

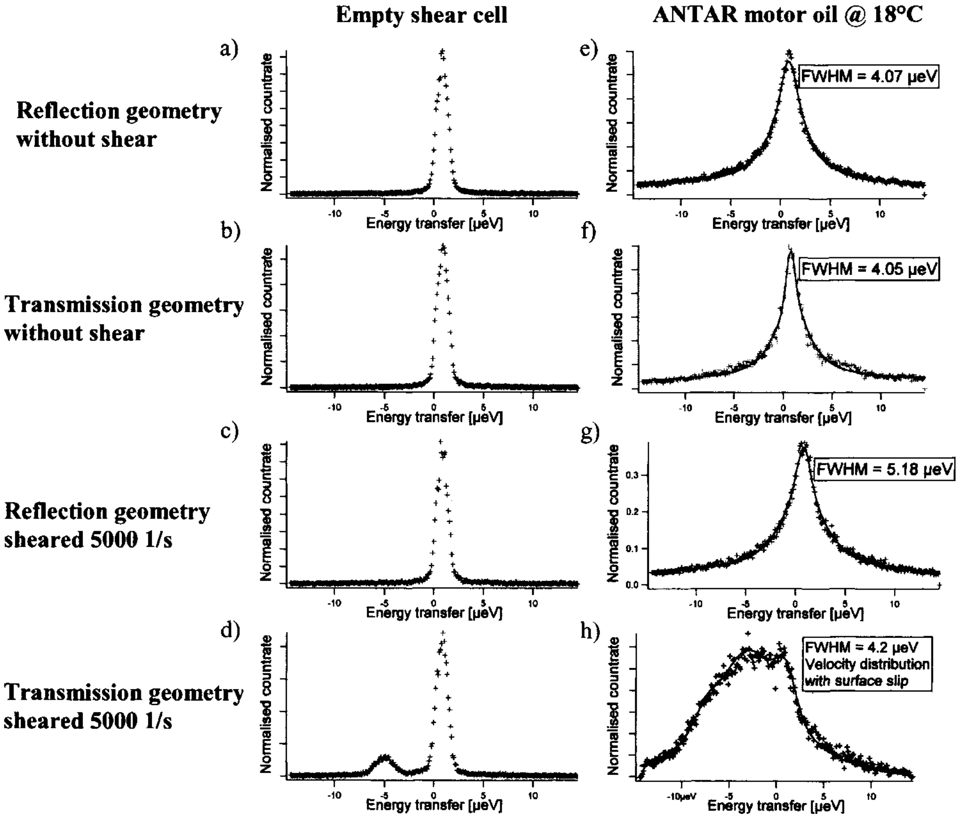

Figure 1. Backscattering spectra obtained at IN16 (ILL). The left side shows spectra of the empty cell with the aluminium discs covered with tape to make them visible for a determination of the instrumental resolution of the individual set-up. The resolution appears identical for the datasets (a, b, c) taken without shear and under shear in reflection geometry. In transmission geometry under shear (d) also inelastic Doppler scattered neutrons from the rotating disc become visible. The right side (e-h) shows data for a sample of the commercial motor oil ANTAR at 18°C. In addition to the information about diffusion processes from quasielastic scattering the inelastic scattering can be directly translated into a macroscopic velocity profile.

The second column of panels (e-h) in figure 1 shows the cell filled with the commercial motor oil ANTAR at a disc spacing of about 0.5 mm in the same scattering geometries as the

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corresponding panels on the left. The elastic line in the upper three pictures (e-g) becomes quasielastic broadened due to diffusion processes within the sample. The two spectra (e, f) taken without shear in transmission and reflection geometry show a similar line width of Γ=4.07 and Γ=4.05 μeV, meaning a diffusion constant of $1{,}57*10^{-5}$ mm^2/s, calculated from Fick's law for random jump diffusion ($D=\Gamma\pi/hQ^2$ with h the Planck constant) . For the sheared sample in reflection geometry (g) the quasielastic line clearly broadens to about Γ=5.18 μeV ($D=2{,}01*10^{-5}$ mm^2/s) indicating an acceleration of the diffusion modes in the direction of the shear gradient in the investigated dynamic region and for a momentum transfer of $Q=1.4$ $Å^{-1}$. In the fourth picture (h) the inelastic scattering reflects directly the velocity distribution of the sample between the discs. To interpret this spectra we fit the data by a scattering law which involves a convolution of the Gaussian instrumental resolution with a Lorenzian line width for the quasielastic scattering. Further a velocity distribution function for the inelastic part of the spectrum is taken into account. Simulations have been carried out to calculate the spectrum for various stepwise linear velocity distributions with different quasielastic line widths. The best fit has been found for a Lorenzian line width of Γ=4.2 μeV, being quite similar to the value found for the sample at rest. This means that diffusion turns from isotropic to anisotropic by the applied external force. The velocity distribution that fits the spectrum best has a lower and an upper limit of 0.15 and 0.70 times the velocity of the rotating disc, respectively, with a linear increase between these two boundary values. This implies a non Newtonian behaviour with large internal friction within the liquid and a weak interaction at the solid-liquid interface, resulting in a pronounced surface slip at a temperature of 18°C.

Neutron diffraction. Figure 2 shows results of an experiment deducted at ADAM, ILL. Rocking curves were taken at different shear rates for two samples with concentrations of 20 ml and 75 ml of graphite particles suspended in 100 ml paraffin with a viscosity of 200 mPas.

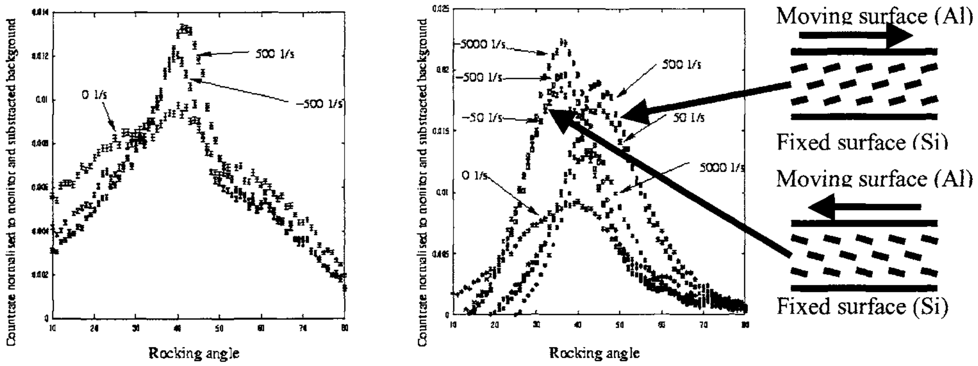

Figure 2. Rocking curves of the 002 Bragg reflection for a suspension of graphite particles (20 ml left side and 75 ml right side in 100 ml paraffin). Under shear the particles begin to order with the c-axis parallel to the shear gradient resulting in an increased intensity. For the 75 ml solution the preferred axis of the ordering of the platelets is tilted by about 5° (shown in the diagrams to the right) for the different directions of the flow, a behaviour that is already indicated from the data of the 20 ml suspension.

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Without shear a constant intensity is expected for any rocking scan as is valid for powder diffraction. Relating to the platelet shape of the graphite particles with a diameter of some 10 microns they tend to align parallel to the surfaces in the narrow gap of 500 microns even without shear. This results is an increased intensity at a rocking angle of 41°. Under shear this intensity becomes further enhanced for the low graphite concentration showing a more pronounced ordering of the particles parallel to the discs. For the higher concentration the maximum of intensity under shear becomes clearly shifted by about 5° demonstrating a tilt angle of the platelets to the discs that depends on the direction of the flowing liquid. For the shear rates measured this tilt angle is constant implying that the particles ordering originates mainly from the confined geometry. This is also supported by the finding that only a slight tilt angle has been observed at low concentration, while higher shear rates result in an increased intensity showing that more particles become involved in the ordering. In addition we note an exceptional behaviour for 5000 1/s with low intensity of the reflection the origin of which is unclear at the moment.

CONCLUSION

It has been shown that neutron scattering is an excellent tool to investigate liquids under flow. With the technique of neutron backscattering for a sample of a commercial motor oil a highly non-linear macroscopic velocity distribution with pronounced surface slip has been found. In addition, within the same measurement it has been possible to address microscopic diffusion modes which become accelerated under shear in direction of the shear gradient whereas the component in direction of the flow remains unchanged. This shows that microscopic diffusion modes become anisotropic under shear. In a further experiment the ordering of macroscopic graphite particles under flow has been observed by monitoring the position of the 002 Bragg peak with neutron diffraction. A tilt angle of about 5° between the normal of the solid-liquid interface and the normal of the graphite platelets has been found for high disperse solutions in paraffin.

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