

Hiroshige Matsuoka  
Graduate Student.

Takahisa Kato  
Associate Professor.

Department of Mechanical Engineering,  
The University of Tokyo,  
7-3-1 Hongo, Bunkyo-ku,  
Tokyo, 113, Japan

# Discrete Nature of Ultrathin Lubrication Film Between Mica Surfaces

*A new apparatus which can measure force and separation between surfaces accurately is developed. Ultrathin fluid lubrication film thickness between mica surfaces is measured using this apparatus. Octamethylcyclotetrasiloxane (OMCTS) is used as a lubricant. As a result of the experiment, it is found that when the film thickness is more than about 10 nm (ten times as large as the molecular diameter of OMCTS), there is good agreement with the conventional continuum fluid lubrication theory (EHL theory). In case of film thickness less than 10 nm, however, it deviates from the theoretical prediction and discretization of film thickness is observed. It is considered that this phenomenon is due to the solvation force (structural force), and that the molecular effect cannot be neglected in such an ultrathin lubrication phenomenon as in this experiment.*

## Introduction

Machine elements are being downsized in recent years with the advance of technology. In accordance with the downsizing, the separation between surfaces that move relative to each other becomes smaller and smaller. For example, in the case of a hard disk drive, the flying height of a magnetic head over a disk surface is approaching a few or a few tens of nanometers and a system in which the head-disk interface is immersed in liquid lubricant instead of air has been considered (de Bruyne and Bogy, 1994; Ikeda and Tago, 1995). Therefore, knowledge concerning the characteristics of lubrication for very thin film thickness will be indispensable as the basis of key-technology in the future (van Alsten and Granick, 1988; Homola et al., 1989; Georges et al., 1993(a), (b)). In this study, we investigate the fluid lubrication film thickness, which is one of the characteristics of lubrication.

It is expected that the conventional lubrication theory in which only artificial load is taken into account must be modified under very thin film thickness or very light load conditions, because naturally generated forces, e.g., the van der Waals force (Lifshitz, 1956) and meniscus force (Orr et al., 1975), will not be negligible. However, there are few theories or experiments related to the lubrication film thickness considering these naturally generated forces. Lubrication film thicknesses more than 10~15 nm have been measured in past studies and it is reported that the measured film thickness in these studies agrees well with conventional lubrication theory (Cooper and Moore, 1994; Johnston et al., 1991). But, in their experiments, the external load is a few tens of Newtons and the naturally generated forces described above are of negligible order. On the contrary, the van der Waals force is not negligible in case of a film thickness less than several nanometers and, hence, precise measurement of lubrication film thickness in that region is important and comparison with the conventional theory is required.

The purpose of this study is to design and make an apparatus that can measure the surface separation and normal force accurately and to measure fluid lubrication film thicknesses less than 10 nm using the new apparatus. Results of experiments are compared with the conventional lubrication theory considering the van der Waals force and the meniscus force.

## Apparatus and Specimens

A new apparatus that the authors developed is shown schematically in Fig. 1. Lubricated surfaces are in a crossed cylinder configuration, which is geometrically equivalent to the configuration between a sphere and a plane (Israelachvili, 1992). Optically polished glass lenses are used as the two cylindrical surfaces, and one of the lenses is supported by a rigid arm, the other by a flexible double cantilever spring. When certain forces act between these surfaces, the cylindrical surface supported by the cantilever spring is displaced, then the normal displacement is measured by a high resolution non-contact capacitive displacement sensor. The resolution of the displacement sensor is 0.7~0.8 nm with the cutoff frequency of 3~5 Hz for 30 seconds. Tangential sliding and normal displacements between the surfaces can be applied by elastic stages 1 and 2, respectively. The structure of the elastic stage is shown in Fig. 2. Smooth sliding of a center solid can be realized by utilizing elastic deformation of four leaf springs caused by pushing the center solid as shown in Fig. 2(b) and the elastic stages are driven by piezoelectric actuator. Displacement of the elastic stage can be measured by four strain gauges glued on the leaf springs shown in Fig. 2. Moving speed of the elastic stage is 0.1~500  $\mu\text{m/s}$ . Microstages 1~3 are for coarse movement, and 4 for translating the displacement sensor. All the microstages can be driven remotely by DC motor. The apparatus is placed on an air suspension table in a clean booth where temperature and relative humidity can be controlled to be constant.

It will be required that surface roughness of the solid be less than 1 nm in order to measure lubrication film thicknesses less than 10 nm. The authors use a mica surface because molecularly smooth surface of large area can be obtained easily by cleaving it. This thin mica sheet is then glued to an optically polished glass lens. Octamethylcyclotetrasiloxane (OMCTS), which is a nonpolar liquid, is used as a lubricant. The OMCTS molecule is spherical, with a diameter of about 1 nm (Horn and Israelachvili, 1981). Properties of mica and OMCTS are listed in Table 1 and 2, respectively.

There are some kinds of rheometers similar to the present apparatus from other studies (Homola et al., 1989; van Alsten and Granick, 1990). Because they use interferometry for measuring thin film thickness a laborious procedure is necessary, such as a thin silver coating on the specimen surface. We use a non-contact capacitive displacement sensor which relieves us from the above work, and nontransparent specimens can be tested.

Contributed by the Tribology Division for publication in the JOURNAL OF TRIBOLOGY. Manuscript received by the Tribology Division May 4, 1995; revised manuscript received October 17, 1995. Associate Technical Editor: C. Cusano.

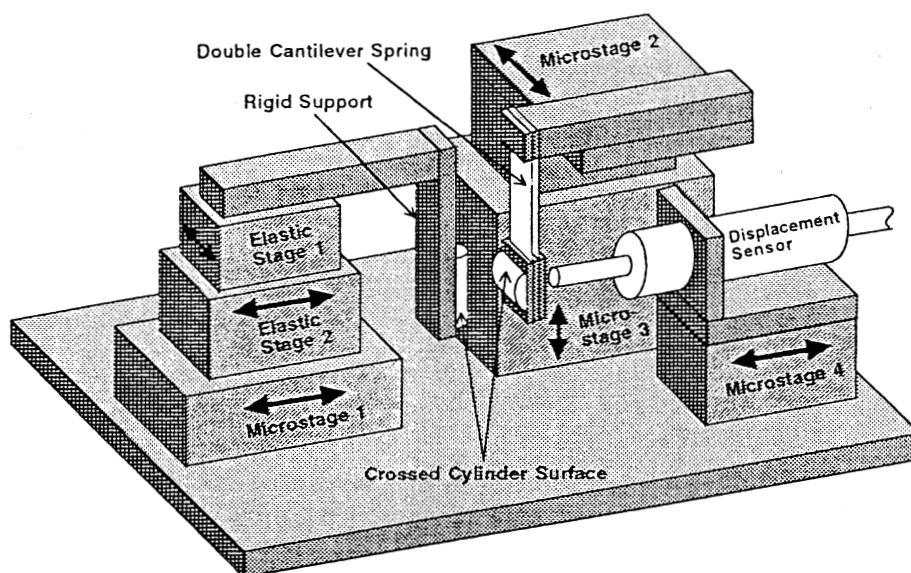


Fig. 1 Apparatus

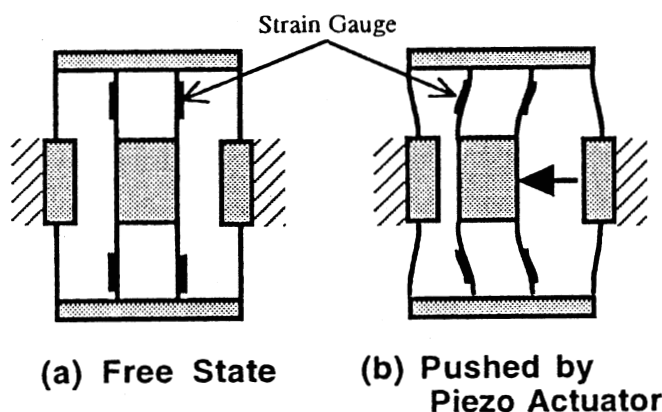


Fig. 2 Elastic stage. (a) Free state; (b) pushed by piezo actuator.

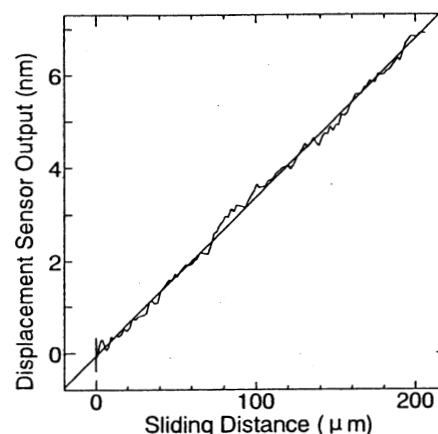


Fig. 3 Dry sliding, showing the small misalignment of the elastic stage

Table 1 Properties of Mica

Chemical formula	$K(Al_2)(Si_3Al)O_{10}(OH)_2^a$
Refractive index, $n$	1.60 <sup>b</sup>
Absorption frequency, $\omega$	$1.9 \times 10^{16}$ rad/s <sup>b</sup>
Dielectric constant, $\epsilon_0$	7.0 <sup>c</sup>
Young's modulus, $E$	34.5 GPa*
Poisson's ratio, $\nu$	0.205*
Roughness	0.048nm(Ra)*, 0.62nm(Rmax)*

a:(Gaines, 1957), b:(Horn and Israelachvili, 1981), c:(Israelachvili, 1992), \* denotes the measured value.

Table 2 Properties of OMCTS

Chemical formula	$[(CH_3)_2SiO]_n^d$
Diameter	1 nm <sup>d</sup>
Refractive index, $n$	1.40 <sup>d</sup>
Absorption frequency, $\omega$	$1.60 \times 10^{16}$ rad/s <sup>d</sup>
Dielectric constant, $\epsilon_0$	2.30 <sup>d</sup>
Contact angle (for mica), $\theta$	7 deg**
Surface energy, $\gamma_L$	19.7 mJ/m <sup>2</sup> <sup>c</sup>
Viscosity, $\eta_0$	2.35 mPa·s <sup>d</sup>

d:(Horn and Israelachvili, 1981), e:(Israelachvili, 1992), \*\* denotes the measured value.

## Experimental Procedure

Square sheets of mica with a size of  $10 \times 10$  mm are cleaved to a thickness of  $10 \sim 30$   $\mu$ m and then glued on the cylindrical optical lenses with cyanoacrylate paste. Curvature radius,  $R$ , of the cylindrical surface is measured by a surface roughness tester ( $R = 10.15 \pm 0.15$  mm). Mass of the cylindrical lens and plates, which are attached to the end of the double cantilever spring (see Fig. 1), is measured by an electric balance. The free vibration frequency of the resulting spring-mass system is measured by FFT analyzer, and the spring constant,  $k_s$ , of the double cantilever spring is calculated from the measured natural frequency of the spring and the mass by a one-degree-of-freedom lumped-mass/spring model ( $k_s = 132.5 \pm 2.5$  N/m). Parallel sliding of the elastic stage 1 is ensured to less than 7 nm (perpendicular to the sliding direction)/100  $\mu$ m (sliding distance). Example of the output from the displacement sensor is shown in Fig. 3, which is obtained by sliding one surface against the other using the elastic stage 1 under dry contact condition. Perfectly parallel sliding of the elastic stage 1 cannot be realized, as shown in Fig. 3, but the output can be approximated by a straight line which passes through the origin of the coordinate axes. The change of load caused by the inclination during the sliding is less than 2  $\mu$ N.

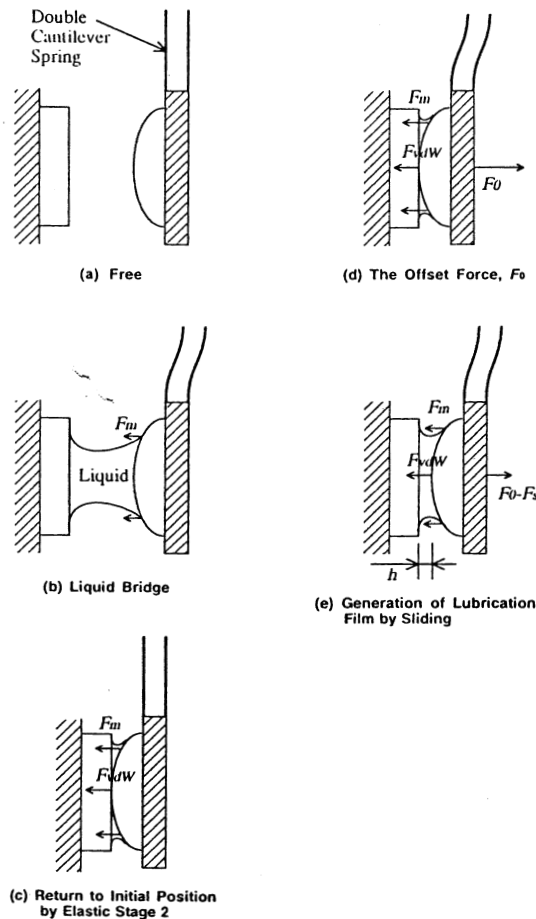


Fig. 4 Procedure of experiment. (a) Free; (b) liquid bridge; (c) return to initial position by elastic stage 2; (d) the offset force,  $F_0$ ; (e) generation of lubrication film by sliding.

Next, the balance of forces is considered carefully. After ensuring parallelism, the two mica surfaces are separated by a small distance (Fig. 4(a)) and a small amount (0.1~0.2 cc) of OMCTS is carefully dropped between the mica surfaces by a syringe. The mica surfaces approach each other by an attractive meniscus force (Fig. 4(b)). The mica surface supported by the double cantilever spring is moved to an initial position in order to remove the deflection of the double cantilever spring using the elastic stage 2 (Fig. 4(c)). Under this condition, two surfaces are attracted to each other by the meniscus force,  $F_m$ , and the van der Waals force,  $F_{vdw}$  and repulsed by the compressive Hertz pressure. Then a pulling force,  $F_0$  is applied by using the double cantilever spring (Fig. 4(d)) in order to cancel  $F_m$  because  $F_m$  is very large compared with  $F_{vdw}$ . Note that  $F_0$  must be smaller than  $F_m$  in order to avoid the surface separation. When the surfaces are slid by the elastic stage 1, the two surfaces separate from each other with film thickness  $h$  because fluid force is generated between the surfaces (Fig. 4(e)). The fluid force is generated by the wedge effect (Cameron, 1966) between mica surfaces and this force is the predominant one in a lubricating film where the lubricant film thickness is relatively large, e.g., larger than 10 nm. The counterbalance of forces is written as:

$$F = F_{vdw} + F_m - (F_0 - F_s) \quad (1)$$

where  $F$  is the fluid force and  $F_0 - F_s$  is pulling force applied by the double cantilever spring and  $F_s = k_s h$ , where  $h$  is the minimum film thickness.

An example of film thickness measurement (output data from the displacement sensor during sliding) is shown in Fig. 5.

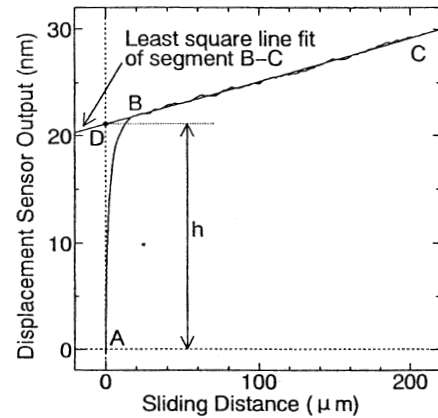


Fig. 5 Positive offset due to the fluid film thickness

While the displacement sensor output increases linearly with sliding distance (B-C in Fig. 5) because sliding is not perfectly parallel as described above, an offset is observed because the fluid film exists between the surfaces. Slightly fluctuating output data B-C are approximated by the first order polynomial least square fitting and the lubrication film thickness,  $h$ , is considered as the segment (D) of the vertical axis by the fitted line because the fitted line passes the origin of the coordinate axes under the dry contact condition as described above (see Fig. 3).

Summarizing the experimental procedure in this study, the film thickness data such as shown in Fig. 5 are measured for a given value of  $F_0$ , then the film thickness,  $h$ , is calculated by least square fitting. The fluid force,  $F$ , is then calculated from expression (1) using the value of the film thickness,  $h$ . This operation is repeated many times and relation between the fluid force,  $F$ , and the film thickness,  $h$ , is obtained.

#### Calculation of $F_{vdw}$ and $F_m$

##### The van der Waals Force Between Macroscopic Bodies.

It is known that the van der Waals force acts between two surfaces when they are separated by a very thin liquid as shown in Fig. 6 (Lifshitz, 1956). Interaction energy per unit area,  $W$ , between planes is given by (Prieve and Russel, 1988):

$$W = A_{131}/12\pi h^2 \quad (2)$$

where  $h$  is the film thickness and  $A_{131}$  is a function of the refractive index,  $n$ , the dielectric constant,  $\epsilon_0$ , and the absorption frequency,  $\omega$ . The van der Waals force,  $F_{vdw}$ , between a sphere and a plane as considered in this study is given as follows (Israelachvili, 1992):

$$F_{vdw} = 2\pi R W = A_{131} R / 6h^2 \quad (3)$$

where  $R$  is curvature radius of the sphere which is equal to that of the cylindrical surface in this study (White, 1983).  $A_{131}$  is often considered as a constant and referred as "Hamaker constant." But, strictly speaking,  $A_{131}$  is not constant and a function of the film thickness,  $h$ , when the film thickness is in

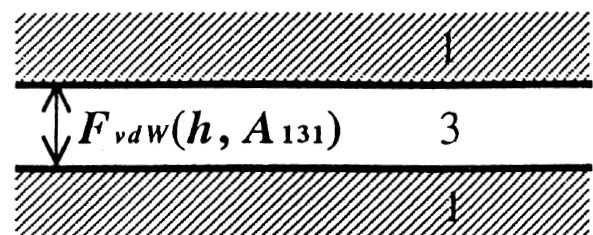


Fig. 6 Interaction between planes

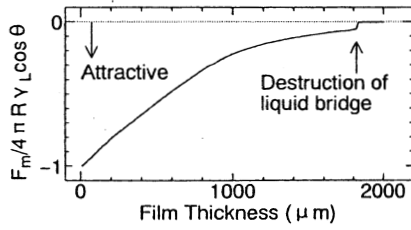


Fig. 7 A change of meniscus force against film thickness

the order of nanometer (see Appendix A). In this condition,  $A_{131}$  is written in a complicated expression as follows (Priev and Russel, 1988; Israelachvili, 1972):

$$A_{131} = -\frac{3}{2} k_B T \sum_{n=0}^{\infty} \int_{r_n}^{\infty} x \{ \ln(1 - \bar{\Delta}_{13}^2 e^{-x}) + \ln(1 - \bar{\Delta}_{13}^2 e^{-x}) \} dx, \quad (4)$$

where

$$\Delta_{jk} = \frac{\epsilon_j s_k - \epsilon_k s_j}{\epsilon_j s_k + \epsilon_k s_j}, \quad \bar{\Delta}_{jk} = \frac{s_k - s_j}{s_k + s_j},$$

$$s_k^2 = x^2 + \left( \frac{2\xi_n h}{c} \right)^2 (\epsilon_k - \epsilon_3)$$

$$r_n = \frac{2h\xi_n \sqrt{\epsilon_3}}{c}, \quad \xi_n = \frac{2\pi n k_B T}{\hbar}, \quad \epsilon_k = \epsilon_k(i\xi_n), \quad \hbar = \frac{h_p}{2\pi}$$

$$\epsilon_k(i\xi_n) = \begin{cases} 1 + \frac{n^2 - 1}{1 + \xi_n^2/\omega_k^2} & (\text{for } \xi_n > 0) \\ \epsilon_{k0} & (\text{for } \xi_n = 0) \end{cases}$$

where  $k_B$  is the Boltzmann constant,  $T$  is the temperature in degrees Kelvin,  $c$  is the speed of light,  $h_p$  is the Planck constant and the prime (') on the summation symbol denotes that the first term is to be multiplied by  $\frac{1}{2}$ .

**The Meniscus Force.** The meniscus force,  $F_m$ , of a sphere-plane interface is given by a function of the film thickness,  $h$  (Israelachvili, 1992; Gao et al., 1995) as in the following expression:

$$F_m = \frac{4\pi R \gamma_L \cos \theta}{1 + ah} \quad (5)$$

where  $\gamma_L$  is the surface energy of liquid,  $\theta$  is the contact angle of liquid for solid surface, and  $a$  is a constant. Change of the meniscus force with film thickness (surface separation) is measured using the apparatus developed in this study and an example of a result is shown in Fig. 7. It is observed that the meniscus force (attractive force) decreases with the film thickness and liquid bridge breaks at some film thickness. It is assumed that the van der Waals force is negligible in this particular experiment because the film thickness is very large. Under this assumption, the constant  $a$  is estimated by using the least square fitting for the film thickness less than  $100 \mu\text{m}$  ( $a = (7.1 \sim 10) \times 10^{-4} (\mu\text{m}^{-1})$ ). Since the film thickness,  $h$ , is smaller than  $0.1 \mu\text{m}$  in the experiment of this study, the value of  $a \times h$  appearing in expression (5) is much smaller than 1 and of negligible order. For simplicity the meniscus force is calculated assuming  $a = 10^{-3} (\mu\text{m}^{-1})$  in the present study.

Comparison between the meniscus force and the van der Waals force is shown in Appendix B.

### Fluid Film Lubrication Regimes

The theoretical relation between fluid force,  $F$ , and lubrication film thickness,  $h$ , is considered in this chapter in order to

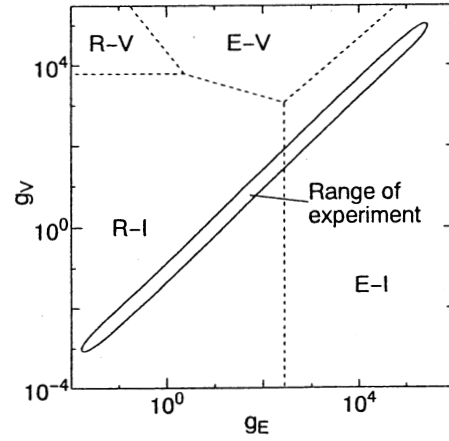


Fig. 8 Lubrication regime in the present study on Hamrock-Dowson diagram

discuss the results of the present experiment. The Reynolds equation and the elasticity equation must be solved simultaneously to obtain the shape of the film thickness between a sphere and a plane, and the relation between  $F$  and  $h$  has been studied and given by simple exponential expression in past studies (Hamrock and Dowson, 1976(a), 1976(b), 1977(a), 1977(b); Evans and Snidle, 1981). There are four regimes classified by lubricating conditions, i.e., load, speed etc. and the applicable expression is different when the lubrication regime is different (Hamrock and Dowson, 1978). Therefore, it is indispensable to know the lubrication regime realized in the present experiment. The four regimes are determined by a combination of properties of the lubricated solid (Rigid or Elastic) and lubricant (Isoviscous or Variableviscous). The Hamrock-Dowson diagram as shown in Fig. 8 has been proposed in order to know the lubrication regime (Hamrock and Dowson, 1978; Esfahanian and Hamrock, 1991). The inside of solid curve drawn in Fig. 8 represents the range of the present experiment. Vertical axis,  $g_v$ , horizontal axis,  $g_E$ , and the parameters in Fig. 8 are as follows:

$$g_E = W^{8/3} U^{-2}, \quad g_v = G W^3 U^{-2},$$

$$W = F/E'R^2,$$

$$G = \alpha E', \quad U = h_0 u / 2E'R,$$

$$R = 10^{-2} (\text{m}), \quad E' = 36.0 (\text{GPa}), \quad \alpha = 10 \sim 30 (\text{GPa}^{-1}),$$

$$h_0 = 2.35 (\text{mPa} \cdot \text{s}), \quad F = 1 \sim 1000 (\mu\text{N}),$$

$$u = 50 \sim 200 (\mu\text{m/s})$$

It is found from Fig. 8 that the lubrication regime in the present experiment is R-I or E-I. The relations between the film thickness,  $h$ , and the fluid force,  $F$ , in the R-I and the E-I regime are written as the following expressions, respectively (Venner and ten Napel, 1992; Hamrock and Dowson, 1978):

$$\text{R-I: } h = 3.55 \times 10^{-5} (\eta_0 u)^2 F^{-2} \quad (6)$$

$$\text{E-I: } h_c = 1.73 \times 10^{-6} (\eta_0 u)^{0.66} F^{-0.21} \quad (7)$$

$$h_{\min} = 1.12 \times 10^{-6} (\eta_0 u)^{0.66} F^{-0.21} \quad (8)$$

where the subscripts  $c$  and  $\min$  denote center film thickness and minimum film thickness, respectively. Note that these formulas (6)–(8) are derived from continuum lubrication theory.

### Results and Discussion

Experimental results and theoretical prediction (formulas (6)–(8)) are shown in Fig. 9. Sliding speed,  $u$ , is  $200 (\mu\text{m/s})$ . The horizontal axis represents the fluid force,  $F$ , and the

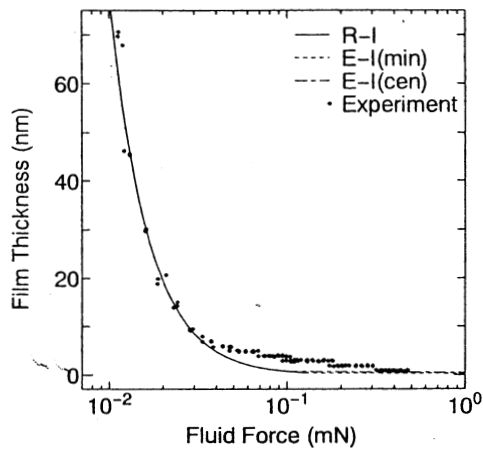


Fig. 9 Comparison between theoretical and experimental film thickness ( $u = 200 \mu\text{m/s}$ )

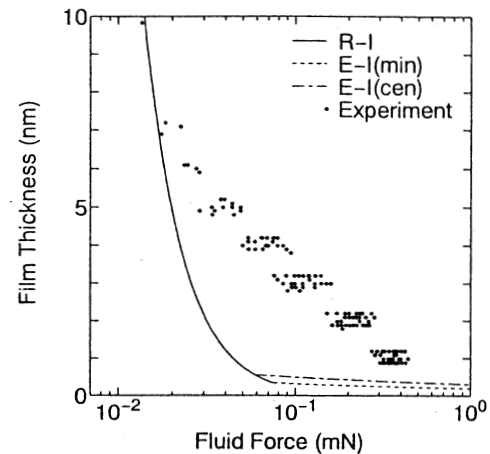


Fig. 11 Ultrathin film thickness ( $u = 100 \mu\text{m/s}$ )

vertical axis represents the lubrication film thickness,  $h$ . It is found that the measured film thickness agrees well with the continuum lubrication theory in R-I regime in the region more than 10 nm. It is reported in past studies that a lubrication film thickness more than 50 nm (Dalmaz, 1978), 15 nm (Johnston et al., 1991), and 10 nm (Cooper and Moore, 1994) agrees well with the theoretical prediction, and the present experiment confirms the past studies. The region of film thickness less than 10 nm in Fig. 9 is enlarged and shown in Fig. 10 in order to observe variations of several nanometers of the film thickness in detail. The film thickness begins to deviate from the theory at about 7–8 nm, and the phenomenon that the film thickness remains constant even if the fluid force increases slightly and the film thickness suddenly jumps down to next stable thickness when the fluid force amounts to some enough large value, i.e., the discrete nature of the film thickness is observed. The interval of the discretized film thickness is about 1 nm, which corresponds roughly to the molecular diameter of OMCTS (see Table 2). Experimental results for the cases that  $u = 100 \mu\text{m/s}$  and  $u = 50 \mu\text{m/s}$  are shown in Fig. 11 and Fig. 12, respectively. The film discretization is again observed. The contact pressure calculated from Hertzian theory is less than 4.5 (MPa) and the shear rates are less than  $2 \times 10^5 \text{ (s}^{-1}\text{)}$  for  $u = 200 \mu\text{m/s}$ , less than  $1 \times 10^5 \text{ (s}^{-1}\text{)}$  for  $u = 100 \mu\text{m/s}$  and less than  $5 \times 10^4 \text{ (s}^{-1}\text{)}$  for  $u = 50 \mu\text{m/s}$ . There is a possibility that the film discretization is an artifact of the measurement device because the resolution of the displacement sensor is 0.7–0.8 nm, but the authors consider the discretization is physical because:

(i) the resolution of the displacement sensor is measured for 30 seconds but the experiment is carried out for several seconds and the short time will result in little thermal drift and finer resolution; (ii) the measured data are averaged by the least square fitting and it will also result in finer resolution; (iii) the value of film thickness is reproducible.

It has been reported that a force peculiar to film thicknesses of several nanometers under static conditions is generated by the influence of the molecular structure of the liquid intervening between solid surfaces (Henderson and Lozada-Cassou, 1986; Horn and Israelachvili, 1981). This peculiar force is called a structural force or a solvation force and cannot be disregarded in case of a film thickness less than 5–10 times as large as the molecular diameter of intervening fluid. The solvation force is oscillatory with the film thickness and the period is almost equal to the molecular diameter of intervening liquid. Fluid molecules in the vicinity of solid surface are arranged stratiformly (Ikeda and Tago, 1995), which can also be the source of the solvation force. Also, in the case of a dynamic condition, it is reported that the squeezed film thickness is discretized due to the solvation force (Chan and Horn, 1985). It is well-known that behavior of the viscosity of a fluid confined to very small spaces is significantly different from bulk viscosity, usually larger, "quantized" by film thickness and dependent on normal pressure (Israelachvili et al., 1988; van Alsten and Granick, 1990; Carson et al., 1992). Larger viscosity results in larger film thickness according to the continuum theory, by which the present results seem to be explained. However, it should be noted that these past experiments were carried out under the condition

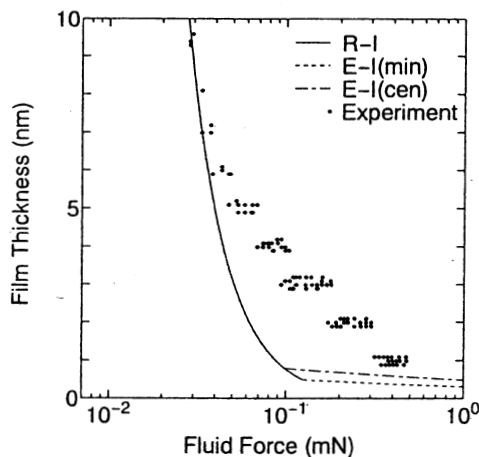


Fig. 10 Enlargement of Fig. 9

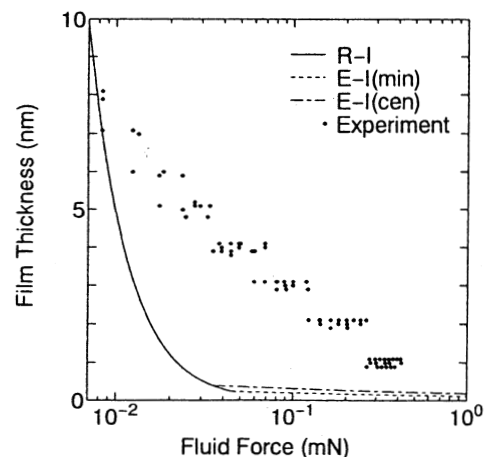


Fig. 12 Ultrathin film thickness ( $u = 50 \mu\text{m/s}$ )

of counterbalance between applied normal force and the solvation force, and it is considered by the above research that this naturally causes the viscosity to be "quantized." Therefore we consider that the discretization of the fluid lubrication film thickness observed in the present work is due to the solvation force.

Recently two groups of researchers (Guanteng and Spikes, 1994; Smeeth et al., 1995) reported that classical EHL theory was obeyed down to less than one nanometer for some fluids and other synthetic fluids gave thicker films than predicted from the EHL theory under very thin film conditions. But they did not observe the film discretization which is observed in the present experiments. The authors consider that this is because (i) surface roughness in their experiments is very large (11 nm) compared with the present experiments ( $R_a = 0.05$  nm,  $R_{max} = 0.6$  nm), and the surface roughness considerably reduces the layering effect of fluid molecules adjacent to solid surface, i.e., it also reduces the solvation force generated by the layering effect (Guanteng and Spikes, 1995; Georges et al., 1993(a); Horn and Israelachvili, 1981); (ii) they used fluids which have chain or branched molecules as lubricants whereas a fluid which has spherical molecules is used in the present study, and the solvation force is difficult to observe when fluids which have chain or branched molecules are used (Christenson et al., 1982; Gee et al., 1990).

Also, it is found from Figs. 10–12 that a higher sliding speed generates a larger fluid force. This is probably due to the conventional hydrodynamic force. The authors consider that the fluid force measured in the present study is mainly composed of the hydrodynamic force and the solvation force.

From these past studies and the present results, in case of the lubrication film thickness in molecular scale, it is considered that the solvation or the structural force caused by the discreteness of fluid molecules and by an arrangement of fluid molecules in the vicinity of solid wall is not negligible compared with the hydrodynamic force caused by the viscosity of the intervening fluid. It is speculated consequently that conventional lubrication theory must not be applied to film thicknesses smaller than several times the molecular diameter of intervening liquid between molecularly smooth surfaces.

## Conclusions

A new apparatus which can measure intersurface force and film thickness accurately is developed. Using this apparatus, the lubrication film thickness between the mica surfaces in a crossed cylinder geometry lubricated by OMCTS is measured. As a result of the experiments, it is found that the lubrication film thickness agrees well with the conventional lubrication theory in case of the film larger than some 10 nm. However, discretization of the film thickness is observed in case of a film thickness of several nanometers. It is considered that the film discretization is due to the solvation force and that continuum lubrication theory cannot be applied in these cases.

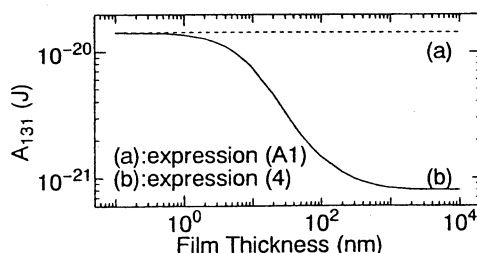


Fig. A1 Variation of  $A_{131}$  with film thickness

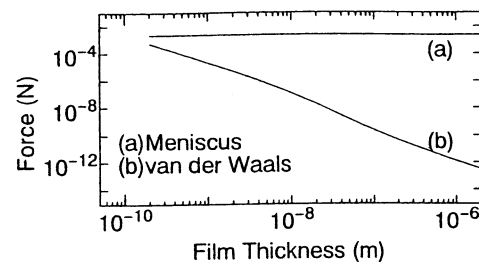


Fig. B1 Meniscus force and van der Waals force

## APPENDIX A

### Discussion About $A_{131}$

$A_{131}$  in expression (3) is written by expression (4) as a function of  $h$ , but  $A_{131}$  is often assumed to be constant and referred to as the Hamaker constant. We discuss here whether  $A_{131}$  can be assumed to be constant or not in case of the film thickness of several nanometers.

$A_{131}$  is approximated by the following expression when the film thickness is very thin ( $h \rightarrow 0$ ) (Horn and Israelachvili, 1981):

$$A_{131} = \frac{3}{4} k_B T \left( \frac{\epsilon_{10} - \epsilon_{30}}{\epsilon_{10} + \epsilon_{30}} \right)^2 + \frac{3\hbar \sqrt{\omega_1 \omega_3}}{64 \epsilon_0^{7/4}} \times \frac{X^2 \bar{\epsilon}_0 + 2X \Delta \epsilon_0 \bar{\epsilon}_0^{1/2} + \Delta \epsilon_0^2 (3 + 2Y)}{[(Y - \sqrt{Y^2 - 1})^{1/2} + (Y + \sqrt{Y^2 - 1})^{1/2}]^3}, \quad (A1)$$

where

$$\bar{\epsilon}_0 = \frac{1}{2} (n_1^2 + n_3^2), \quad X = \frac{\omega_1}{\omega_3} (n_1^2 - 1) - \frac{\omega_3}{\omega_1} (n_3^2 - 1)$$

$$\Delta \epsilon_0 = n_1^2 - n_3^2, \quad Y = \frac{1}{4\epsilon_0^{1/2}} \left\{ \frac{\omega_1}{\omega_3} (n_1^2 + 1) + \frac{\omega_3}{\omega_1} (n_3^2 + 1) \right\}$$

While this expression includes material parameters, but is not a function of the film thickness,  $h$ , any more and regarded as constant. Figure A1 shows the value of  $A_{131}$  given by expression (A1) and numerical solution of expression (4) for mica-OMCTS-mica system. It is found that  $A_{131}$  can be considered constant only when the film thickness is less than about 1 nm. Hence, the approximate expression (A1) is not applicable and expression (4) must be used to calculate the van der Waals force,  $F_{vdw}$ , in case of the film thickness in several nanometers.

## APPENDIX B

### Comparison Between $F_m$ and $F_{vdw}$

In order to know the relative dimension of two forces, the meniscus force is compared with the van der Waals force and it is shown in Fig. B1. The meniscus force is very large, hence the force canceling it,  $F_0$ , is very important in this experiment. The proportion of  $F_{vdw}$  to the fluid force,  $F$ , is less than 10 percent but  $F_{vdw}$  is necessary to the accurate calculation of the fluid force,  $F$ .

## References

- Cameron, A., 1966, *The Principles of Lubrication*, Longmans.
- Carson, G., Hu, H.-W., and Granick, S., 1992, "Molecular Tribology of Fluid Lubrication: Shear Thinning," *STLE Tribology Transactions*, Vol. 35, pp. 405–410.
- Chan, D. Y. C., and Horn, R. G., 1985, "The Drainage of Thin Liquid Films Between Solid Surfaces," *Journal of Chemical Physics*, Vol. 83, pp. 5311–5324.
- Christenson, H. K., Horn, R. G., and Israelachvili, J. N., 1982, "Measurement of Forces Due to Structure in Hydrocarbon Liquids," *Journal of Colloid and Interface Science*, Vol. 88, pp. 79–88.



- Cooper, D., and Moore, A. J., 1994, "Application of the Ultra-Thin Elastohydrodynamic Oil Film Thickness Technique to the Study of Automotive Engine Oils," *Wear*, Vol. 175, pp. 93–105.
- Dalmaz, G., 1978, "Film Thickness and Traction Measurements in Small Elastohydrodynamic Elliptical Contacts," *Proceedings of the 5th Leeds-Lyon Symposium on Tribology*, pp. 71–80.
- de Bruyne, F. A., and Bogoy, D. B., 1994, "Numerical Simulation of the Lubrication of the Head-Disk Interface Using a Non-Newtonian Fluid," *ASME JOURNAL OF TRIBOLOGY*, Vol. 116, pp. 541–548.
- Esfahanian, M., and Hamrock, B. J., 1991, "Fluid-Film Lubrication Regimes Revisited," *STLE Tribology Transactions*, Vol. 34, pp. 628–632.
- Evans, H. P., and Snidle, R. W., 1981, "The Isothermal Elastohydrodynamic Lubrication of Spheres," *ASME JOURNAL OF LUBRICATION TECHNOLOGY*, Vol. 103, pp. 547–557.
- Gaines, Jr., G. L., 1957, "The Ion-Exchange Properties of Muscovite Mica," *Journal of Physical Chemistry*, Vol. 61, pp. 1408–1413.
- Gao, C., Tian, X., and Bhushan, B., 1995, "A Meniscus Model for Optimization of Texturing and Liquid Lubrication of Magnetic Thin-Film Rigid Disks," *STLE Tribology Transactions*, Vol. 38, pp. 201–212.
- Gee, M. L., McGuiggan, P. M., Israelachvili, J. N., and Homola, A. M., 1990, "Liquid to solid like transition of molecularly thin films under shear," *Journal of Chemical Physics*, Vol. 93, pp. 1895–1906.
- Georges, J. M., Millot, S., Loubet, J. L., and Tonk, A., 1993(a), "Drainage of thin liquid films between relatively smooth surfaces," *Journal of Chemical Physics*, Vol. 98, pp. 7345–7360.
- Georges, J. M., Millot, S., Loubet, J. L., Tonk, A., and Mazuyer, D., 1993(b), "Surface Roughness and Squeezed Films at Molecular Level," *Proceedings of the 19th Leeds-Lyon Symposium on Tribology*, pp. 443–452.
- Guanteng, G., and Spikes, H. A., 1994, "Behavior of Lubricants in the Mixed Elastohydrodynamic Regime," presented at the 21st Leeds-Lyon Symposium on Tribology, *Lubricants and Lubrication*.
- Guanteng, G., and Spikes, H. A., 1995, "Boundary Film Formation by Lubricant Base Fluids," presented at the 50th STLE annual meeting, will be published in *Tribology Transactions*.
- Hamrock, B. J., and Dowson, D., 1976(a), "Isothermal Elastohydrodynamic Lubrication of Point Contacts Part I-Theoretical Formulation," *ASME JOURNAL OF LUBRICATION TECHNOLOGY*, Vol. 98, pp. 223–229.
- Hamrock, B. J., and Dowson, D., 1976(b), "Isothermal Elastohydrodynamic Lubrication of Point Contacts Part II-Ellipticity Parameter Results," *ASME JOURNAL OF LUBRICATION TECHNOLOGY*, Vol. 98, pp. 375–383.
- Hamrock, B. J., and Dowson, D., 1977(a), "Isothermal Elastohydrodynamic Lubrication of Point Contacts Part III-Fully Flooded Results," *ASME JOURNAL OF LUBRICATION TECHNOLOGY*, Vol. 99, pp. 264–276.
- Hamrock, B. J., and Dowson, D., 1977(b), "Isothermal Elastohydrodynamic Lubrication of Point Contacts Part IV-Starvation Results," *ASME JOURNAL OF LUBRICATION TECHNOLOGY*, Vol. 99, pp. 15–23.
- Hamrock, B. J., and Dowson, D., 1978, "Minimum Film Thickness in Elliptical Contacts for Different Regimes of Fluid Film Lubrication," *Proceedings of the 5th Leeds-Lyon Symposium on Tribology*, pp. 22–27.
- Henderson, D., and Lozada-Cassou, M., 1986, "A Simple Theory for the Force between Spheres Immersed in a Fluid," *Journal of Colloid and Interface Science*, Vol. 114, pp. 180–183.
- Homola, A. M., Israelachvili, J. N., Gee, M. L., and McGuiggan, P. M., 1989, "Measurement of and Relation Between the Adhesion and Friction of Two Surfaces Separated by Molecularly Thin Liquid Films," *ASME JOURNAL OF TRIBOLOGY*, Vol. 111, pp. 675–682.
- Horn, R. G., and Israelachvili, J. N., 1981, "Direct Measurement of Structural Forces between Two Surfaces in a Nonpolar Liquid," *Journal of Chemical Physics*, Vol. 75, pp. 1400–1411.
- Ikeda, Y., and Tago, K., 1995, "Tribological Study on Molecularly Thin Films Using Non-Equilibrium Molecular Dynamics," *JAST Journal of Japanese Society of Tribologists*, Vol. 40, pp. 253–259.
- Israelachvili, J. N., 1972, "The Calculation of van der Waals Dispersion Forces Between Macroscopic Bodies," *Proceedings of the Royal Society of London, Series A*, Vol. 331, pp. 39–55.
- Israelachvili, J. N., McGuiggan, P. M., and Homola, A. M., 1988, "Dynamic Properties of Molecularly Thin Liquid Films," *Science*, Vol. 240, pp. 189–191.
- Israelachvili, J. N., 1992, *Intermolecular and Surface Forces*, 2nd edition, Academic Press.
- Johnston, G. J., Wayte, R., and Spikes, H. A., 1991, "The Measurement and Study of Very Thin Lubricant Films in Concentrated Contacts," *STLE Tribology Transactions*, Vol. 34, pp. 187–194.
- Lifshitz, E. M., 1956, "The Theory of Molecular Attractive Forces between Solids," *Soviet Physics, JETP*, Vol. 2, pp. 73–83.
- Orr, F. M., Scriven, L. E., and Rivas, A. P., 1975, "Pendular Rings Between Solids: Meniscus Properties and Capillary Force," *Journal of Fluid Mechanics*, Vol. 67, pp. 723–742.
- Prieve, D. C., and Russel, W. B., 1988, "Simplified Predictions of Hamaker Constants from Lifshitz Theory," *Journal of Colloid and Interface Science*, Vol. 125, pp. 1–13.
- Smeeth, M., Spikes, H. A., and Günsel, S., 1995, "The Formation of Viscous Surface Films by Polymer Solutions: Boundary or Elastohydrodynamic Lubrication?," presented at the 50th STLE annual meeting, will be published in *Tribology Transactions*.
- van Alsten, J., and Granick, S., 1988, "Molecular Tribometry of Ultrathin Liquid Films," *Physical Review Letters*, Vol. 61, pp. 2570–2573.
- van Alsten, J., and Granick, S., 1990, "Tribology Studied Using Atomically Smooth Surfaces," *STLE Tribology Transactions*, Vol. 33, pp. 436–446.
- Venner, C. H., and ten Napel, W. E., 1992, "Multilevel Solution of the Elastohydrodynamically Lubricated Circular Contact Problem—Part 2: Smooth Surface Results," *Wear*, Vol. 152, pp. 369–381.
- White, L. R., 1983, "On the Deryaguin Approximation for the Interaction of Macrobodies," *Journal of Colloid and Interface Science*, Vol. 95, pp. 286–288.