

# New achievements in superlubricity from International Workshop on Superlubricity: Fundamental and Applications

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**Abstract:** Since the term “superlubricity” was put forward at the beginning of 1990s, it has become one of the hottest researches in tribology due to it being close linked to the energy problems. Recently, the International Workshop on “Superlubricity: Fundamental and Applications” was successfully held on 19–20 October 2015 in Beijing, which has attracted many researchers in this field. The recent scientific results in both solid superlubricity and liquid superlubricity have been presented according to these invited wonderful lectures and posters. In the communication, we gave an introduction to the Workshop on Superlubricity, and also summarized the new achievements of superlubricity during recent years according to these reports. Finally, the problems of superlubricity mechanism and the future development direction of superlubricity are discussed.

**Keywords:** friction; solid superlubricity; liquid superlubricity

## 1 Introduction

Superlubricity is a regime of motion in which the friction between two contact surfaces approaches or reaches zero. Because of the ultra-low friction force acting in superlubricity, the losses in energy and material caused by friction and wear in mechanical systems become very small. Therefore, investigations on superlubricity technologies and mechanisms are significant in saving energy in both industrial and daily life applications. Superlubricity has attracted much attention from researchers in tribology, physics, chemistry, and materials; the research may lead to revolutionary new technologies with extremely high performance, long life, and low energy consumption [1].

The International Workshop “Superlubricity: Fundamental and Applications” was held on 19–20 October 2015 in Beijing. The workshop was organized by Professors Quanshui Zheng from XIN Center of Tsinghua University, Michael Urbakh from Tel Aviv University, and Jianbin Luo from the State Key

Laboratory of Tribology, Tsinghua University. The workshop was intended to discuss recent scientific results from studies on different aspects of superlubricity in nano- to macroscale systems, including dry and lubricated frictional contact states, and to tabulate unresolved problems for future investigation. Almost 20 keynote lectures and 20 posters were invited, demonstrating the breadth and timeliness of the subject. Approximately 200 experts, teachers, and graduate students from America, China, Germany, Israel, Italy, Japan, the Netherlands, Sweden, and Switzerland attended intensive discussions and interactions during the workshop. The keynote lectures focused on recent achievements in solid superlubricity, liquid superlubricity, and theoretical simulations of structural lubricity and the potential applications of these phenomena. These achievements demonstrate significant development in superlubricity.

## 2 Solid superlubricity

It was first put forward that the friction force approaches zero when the two contacting crystal planes are sliding

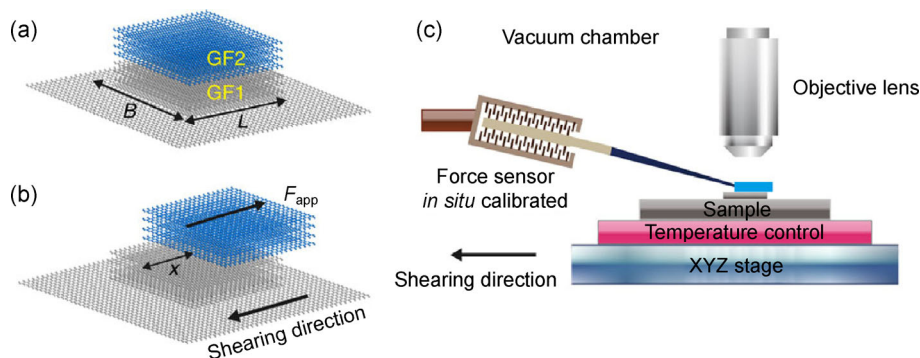
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in incommensurate direction in the early 1990s [2]. After this discovery, many typical materials were found to have superlubricity properties, including graphite, diamond-like carbon (DLC), fullerene-like carbon,  $\text{MoS}_2$ , and graphene. Prof. Quanshui Zheng from Tsinghua University, Prof. Junyan Zhang from the Lanzhou Institute of Chemical Physics, Dr. Anirudha V. Sumant from the Argonne National Laboratory, and Dr. Elad Koren from IBM Research–Zürich presented detailed elaborations and discussions on new behaviors of solid superlubricity from the meso-to macroscale.

Prof. Quanshui Zheng gave a presentation on understanding structural superlubricity by performing experiments in graphite, the challenges met, and the possible applications of the results. Graphite is among the most important solid lubricants because it has a lamellar structure with very strong covalent bonding within the basal plane, but extremely weak van der Waals bonding between planes. Prof. Zheng noted that the biggest challenge to realizing superlubricity at larger scales under ambient conditions is obtaining two atomically flat surfaces at larger scales that can make contact with each other without absorptions, contaminants, or both between them. The self-retracting motion phenomenon in graphite was first observed by Zheng et al.; the property provides a novel solution to this material challenge [3]. Based on the novel property of self-retracting motion, Prof. Zheng revisited and shared his group’s experimental observations from the last ten years, the understandings reached from the results, and the remaining points of confusion [4–6]. The basal plane cleavage energy (CE)

of graphite was identified as a crucial material parameter for understanding many of the unusual properties of graphite, graphene, and carbon nanotubes. Prof. Zheng’s group first reported accurate experimental results for the CE of incommensurate graphite on the basal plane, using a novel method based on the self-retraction phenomenon, as shown in the Fig. 1 [7]. These experimental measurements realized a novel application of superlubricity in graphite.

Prof. Junyan Zhang spoke on achieving the engineering ultra-low friction nature of fullerene-like-structured hydrogenated-carbon thin films. He proposed the concept that fullerene-like carbon (FL-C) nanostructures can extend the inert or vacuum ultra-low friction behaviors of hydrogenated carbon (C:H) films into open or humid air, making further engineering applications practical [8]. The FL-C structures endow the C:H films with high hardness and elasticity, leading to extremely tough, elastic, and compliant films much like “superhard rubbers”, which render the films capable of dissipating contact pressure over large areas by elastic deformation by bond-angle deflection, rather than bond breakage [9]. More importantly, Prof. Zhang’s group found that the surfaces of the structural films undergo gradual transformations into more rich and stable FL-C structures, because the number of pentagonal and heptagonal carbon rings, in addition to hexagonal graphene rings, increases as a result of the thermal and strain effects from repeated friction [10]. The ultra-low friction mechanism from the *in-situ* production of richer FL-C structures in sliding contact provides



**Fig. 1** Illustration of the cleavage energy (CE) measurement. (a) Graphite sample. (b) The CE is measured by shearing the lower flake relative to the upper one in the superlubricated state. (c) Schematic of the experimental setup to shear the sample using an XYZ stage, measuring the shear force  $F_{app}$  using an *in situ* calibrated and fixed micro-force sensor under controlled temperature and vacuum atmosphere (Reproduced with permission from Ref. [7], Copyright 2015, Nature Publishing Group).

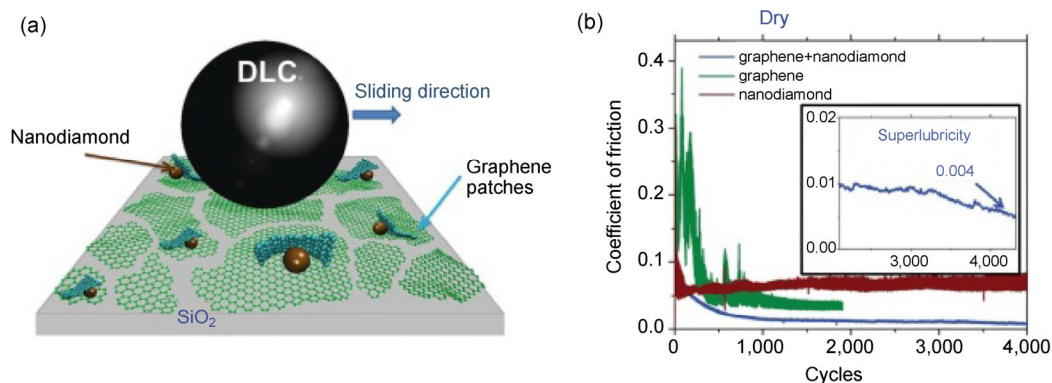
the possibility of new designs further optimizing the tribological performances of the films by adjustment with hydrogen or argon. This simple route to achieve low friction and long wear life for FL-C films in open air is feasible for most frictional parts of an engine.

Graphene has shown the ability to substantially decrease friction and wear under specific conditions at both nano- and macroscales. However, sustained macroscale superlubricity, particularly at engineering scales, has yet to be demonstrated. Dr. Anirudha V. Sumant reported and discussed the mechanism of macroscale superlubricity in graphene-nanodiamond ensembles. His group used experimental studies to demonstrate stable macroscale superlubricity when sliding a graphene-coated surface against a DLC-coated counter face. Meanwhile, the underlying causes and mechanisms of superlubricity were revealed. Graphene patches at a sliding interface wrapped around the nanodiamonds to form nanoscrolls with reduced contact areas, which slid against the DLC surface, achieving an incommensurate contact, which in turn substantially reduced the coefficient of friction to 0.004, as shown in Fig. 2 [11].

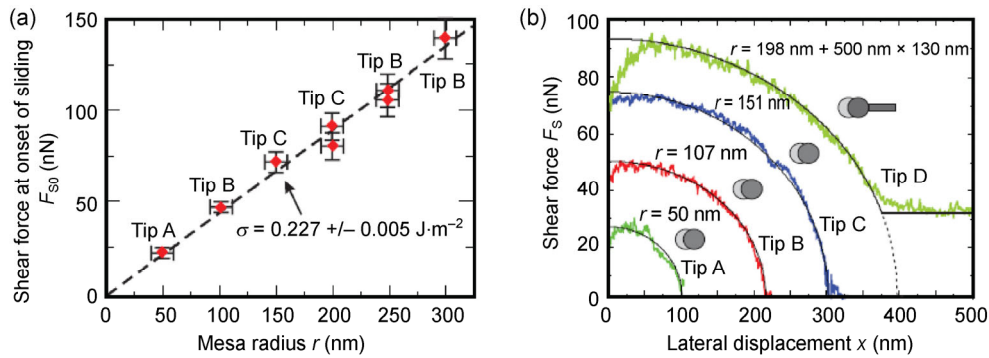
The weak interlayer binding in two-dimensional (2D) layered materials, such as graphite, promotes low friction properties, enabling actuation solely driven by interfacial forces. However, the mechanism of the low friction as well as accurate measurements of the adhesion forces governing the overall mechanical stability has remained elusive to date. Dr. Elad Koren presented on the adhesion and friction forces in mesoscale graphite sliding contacts, reporting the

direct mechanical measurement of line tension and friction forces acting in sheared mesoscale graphite structures. His group exploited the unique nano-positioning accuracy and force sensitivity of scanning-probe microscopy to repeatedly shear individual nano-sized graphitic mesa structures, measuring the applied shear forces during the mechanical exfoliation of the graphite along a single basal glide plane [12]. The adhesive surface free energy ( $0.227 \pm 0.005 \text{ J}\cdot\text{m}^{-2}$ ) was measured in excellent agreement with theoretical models, as shown in Fig. 3. In addition, he showed that the friction is fundamentally stochastic in nature and due to the interaction between the non-commensurate interface lattices. The effect originates from the cancellation of forces acting on the interfacial atoms, excepting those at the periphery. The exponent  $\gamma$  in the scaling equation  $F_{\text{fr}} = A^\gamma$  can assume values in the range from 0 to 0.5, depending on the type of lattice mismatch and the shape of the sliding object. Recently, fractional scaling was experimentally confirmed by measuring the sliding friction forces of amorphous antimony and single-crystal gold islands on a graphite surface [13]. Koren's group found that  $\gamma = 0.35 \pm 0.05$ , in agreement with the latter experiments, suggesting that the friction force originates from interactions between the rotationally misaligned graphite lattices at the sliding interface [14].

In addition to experimental studies on solid superlubricity, many works have explored solid superlubricity by theoretical simulations. Professors Annalisa Fasolino from Radboud University Nijmegen, Motohisa Hirano from Hosei University, Erio Tosatti from SISSA/



**Fig. 2** Experimental demonstration of the superlubricity regime. (a) Schematic (not to scale) of the superlubricity test. (b) The coefficient of friction (COF) for a DLC ball sliding in a dry nitrogen environment against (i) graphene-plus-nanodiamonds (superlubricity state with COF  $\sim 0.004 \pm 0.002$ ), (ii) graphene alone (COF  $\sim 0.04 \pm 0.01$ ), and (iii) nanodiamond alone (COF  $\sim 0.07 \pm 0.01$ ). Inset depicts a plot for superlubricity. (Reproduced with permission from Ref. [11], Copyright 2015, American Association for the Advancement of Science).

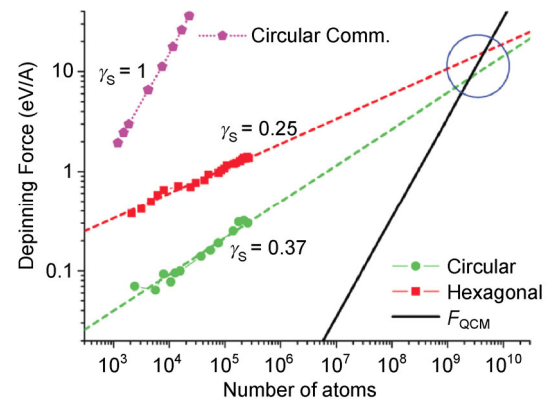


**Fig. 3** Conservative forces due to adhesion energy. (a) Shear force at onset of mesa sliding  $F_{S0}$  as a function of the radius of the cylindrical mesas. (b) Measured shear force  $F_S$  as a function of the lateral displacement  $x$  of the mobile top mesa section for different mesa structures and different cantilevers (Reproduced with permission from Ref. [14], Copyright 2015, American Association for the Advancement of Science).

ICTP, Michael Urbakh from Tel Aviv University, Nainchang Yeh from Cal Tech and Jianbin Luo from Tsinghua University reported and discussed new developments in theory and simulation for solid-structure superlubricity. Prof. Jianbin Luo introduced the theoretical attempts to solve the problem of solid superlubricity through molecular dynamics simulations. By introducing large intrinsic lattice mismatches between fluorographene (FG) and  $\text{MoS}_2$  monolayers, the interlayer friction could be reduced to 1/40 of that in FG/FG and 1/160 of that in  $\text{MoS}_2/\text{MoS}_2$  layered structures. The superlubricity, regardless of the interfacial orientation in heterostructures, is attributed to the formation of Moiré patterns and perpetual interfacial mismatches, which lead to an ultra-smooth potential energy surface and the cancellation of lateral forces [15]. He also introduced an experimental method to measure the friction between 2D materials by using a graphene-coated microsphere probe, where stable superlubricity under high contact pressures can be achieved.

Prof. Erio Tosatti from SISSA/ICTP reviewed some surprising results of simulations in the sliding of incommensurate systems. Mismatches at the interface of two periodic crystals can promote free sliding, a situation called structural lubricity or superlubricity, if the sliding crystals are sufficiently hard in comparison to their mutual corrugation. Aubry's simulations have suggested that the Aubry transition also exists in more realistic 2D cases; however, his group found the transition to be abrupt and of the first order [16]. A second surprise, once again appearing in simulation,

was the necessary imperfect equilibrium angular alignment of an incommensurate monolayer with a 2D periodic substrate, because the two materials generally acquire a small, but finite, misalignment angle [17]. A third surprise, recently clarified in the sliding of crystalline rare-gas solid islands or gold clusters on incommensurate crystal surfaces, is the presence of edge-friction. The superlubric sliding of an island of area  $A$  implies that friction grows as  $A^\gamma$ , where  $\gamma < 1$ . It was demonstrated by simulation that rare gas islands on a metal indeed yield  $0.25 < \gamma < 0.37$ , depending on the shape of the edge, as shown in Fig. 4 [18]. A fourth result emerged in a recently proposed frictional emulator, consisting of short cold ion N-member chains sliding in an optical lattice [19]. The surprising result was the surprising speed with



**Fig. 4** Growth of static friction of an incommensurate rare gas monolayer island with increasing island size. The sublinear scaling indicates superlubricity; residual friction is due to the island edge (Reproduced with permission from Ref. [18], Copyright 2014, Royal Society of Chemistry).



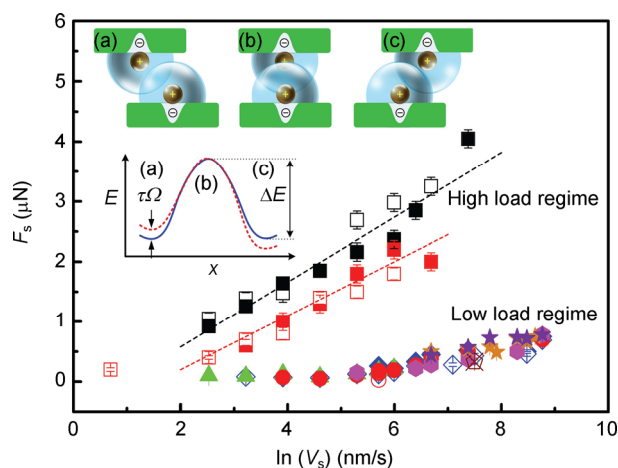
which the superlubric drop of friction developed as  $N$  increased from the value of one. The drop in “atomic” stick-slip friction with increasing heterogeneity appears in chains as short as  $N = 3$  ions [20].

### 3 Liquid superlubricity

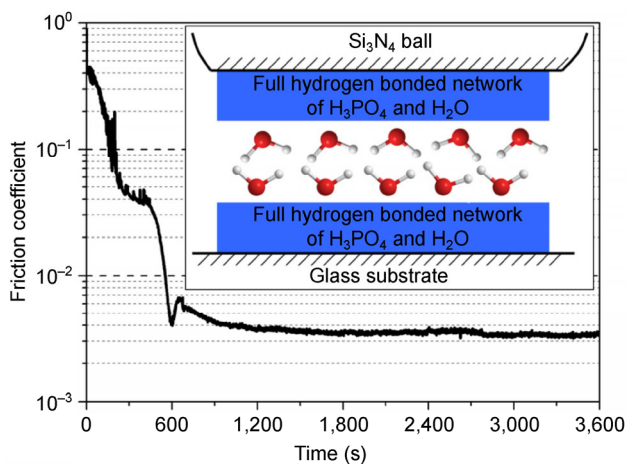
In addition to solid superlubricity, surfaces (or friction systems) lubricated with several liquid materials also have displayed superlubricity properties, such as ceramic materials with water, polymer brushes, phosphoric acid solutions, glycerol or polyhydric alcohol solutions with acid, and some kinds of polysaccharide mucilage from plants. Professors Jacob Klein from the Weizmann Institute of Science, Jianbin Luo from Tsinghua University, and Koshi Adachi from Tohoku University provided detailed reports and discussions on the mechanism of liquid superlubricity at both the nano- and macroscale. Prof. Jacob Klein spoke on superlubricity behavior in aqueous media at nano-scale. The aqueous media could be salt solutions or charged polymer brushes [21, 22]. The main mechanism is attributed to hydration lubrication, in which the hydration shells surrounding charges act as lubricating elements in boundary layers. It is also one of the most important lubrication models in the living organisms, such as joint lubrication and eye lubrication. However, some of the basic properties of hydration lubrication, such as the frictional energy dissipation, remain unclarified despite decades of study. In all relevant studies to date, the measured friction was dominated by other dissipation pathways, such as polymer chains disentangling from each other or the distortion of sheared liposomes [23]. Recently, Klein’s group overcame these limitations by trapping hydrated ions in the gap between molecularly smooth surfaces, and measuring the friction between the surfaces as they slid at pressures and shear rates one or two orders of magnitude higher than those previously achieved. This allowed the clear observation of different dissipation regimes, and enabled the isolation of frictional dissipation arising from the shear of the sub-nanometer hydration shells themselves, intrinsic to all hydration lubrication processes, from any system-dependent dissipation mechanisms, as shown in Fig. 5 [24]. The experiment suggested that frictional

forces even in such simple systems can be dominated by rate-activated pathways. They also found that the viscosity of the sub-nanometer hydration layers could be some 250 times higher than the viscosity of either bulk water or similarly confined non-hydration water. This shed great insight on the causes of hydration lubrication, with implications for both aqueous boundary lubricants and bio-lubrication.

Prof. Jianbin Luo reviewed the history of liquid superlubricity since its discovery and gave a summary of the progress in researching liquid superlubricity at the State Key Laboratory of Tribology. In 2008, Luo’s group first found that yoghurt can achieve superlubricity; however, they later found that the superlubricity results were incorrect because measuring error, which is very sensitive to superlubricity, was not considered in the test process [25]. In 2011, the superlubricity of phosphoric acid solution with an ultra-low friction coefficient of 0.004 was discovered by Luo’s group, as shown in Fig. 6 [26]. They noted that hydrogen ions can be adsorbed onto the friction surfaces by tribochemical reactions and attributed the superlubricity mechanism to the formation of hydrogen-bonded network [27]. They also built a theoretical model of the superlubricity of phosphoric acid by molecular dynamics simulation methods [28]. Another typical material found to have superlubricity is glycerol mixture. In 2008, Ma et al. found that



**Fig. 5** Variation of shear forces with sliding velocity. Variation of shear force  $F_s$  with sliding velocity  $v_s$ , in both high- and low-load regimes (plotted as  $F_s$  versus  $\ln(v_s)$ ). Shown are data at two high loads (black squares:  $F_n = 8$  mN; red squares:  $F_n = 6$  mN) and at several low loads (Reproduced with permission from Ref. [24], Copyright 2015, Nature Publishing Group).



**Fig. 6** Friction coefficient with time under the lubrication of aqueous  $\text{H}_3\text{PO}_4$  solution (pH = 1.5). Inset: lubrication model of superlubricity of  $\text{H}_3\text{PO}_4$  solution (Reproduced with permission from Ref. [26], Copyright 2011, American Chemical Society).

superlubricity could be obtained with the lubrication of a mixture of glycerol and boric acid after a running-in process of a few minutes [29]. After that, Li et al. studied the superlubricity phenomenon of glycerol. They found that all acidic solutions mixed with polyhydroxy alcohol solutions could achieve superlubricity [30]. The ultra-low friction was closely related to the pH value of acid and the concentration of polyhydroxy alcohol. Recently, it was found that the superlubricity of oil-based lubricants could also be achieved by running-in with an acidic solution [31]. The superlubricity mechanism is attributed to the formation of a hydrodynamic film between two friction surfaces by the function of hydrogen ions. The work confirmed that liquid superlubricity could be achieved not only by surface chemical actions, but also by hydrodynamic lubrication under special conditions. Finally, they established a liquid superlubricity region based on the contact pressure and pressure viscosity coefficient [32].

Prof. Koshi Adachi reported that liquid superlubricity could be achieved by controlling the running-in process to create a nano-interface. He first reviewed the mechanism of super-low friction of  $\text{CN}_x/\text{CN}_x$  in inert gases, and noted that the running-in process at the beginning of sliding is important for achieving superlubricity. In addition, he also investigated the superlubricity behavior of  $\text{Si}_3\text{N}_4/\text{CN}_x$  with water as lubricant. A running-in process was required to form very smooth interfaces; meantime, a silica layer was

formed by tribochemical reactions. The superlubricity mechanism is attributed to the formation of the double electrical layer on the silica layer and a hydrodynamic film between the two friction surfaces [33]. The running-in process was found to be able to change the original surface free energy and surface roughness, which could affect the final superlubricity properties. The relationship between superlubricity and surface texture was also investigated, showing that the load capacity of liquid superlubricity could be improved greatly by the introduction of special surface textures [34].

#### 4 Concluding remarks and perspectives

From these reports above, studies in solid superlubricity have focused on the theoretical mechanism of superlubricity. Atomic force microscope (AFM) is widely used by many researchers as an effective apparatus to investigate the mechanisms of solid superlubricity. Many interesting phenomena produced by solid superlubricity, such as self-retraction, pulling of molecular chains, and manipulation of nanoparticles, can be observed directly by AFM. At present, solid superlubricity can be achieved at different scales. For example, graphene or  $\text{MoS}_2$  show superlubricity at the nanoscale, while graphite or DLC do so at the macroscale. The mechanism is mainly attributed to incommensurate structures or weak interfacial interactions. Although the theory is perfect, the types of solid superlubricity materials are limited, and only a few typical materials are known to show superlubricity properties. The existence of other solid materials with superlubricity properties remains unclear; the identification of such materials deserves our speculation and study. Moreover, the application of solid superlubricity under the present technology is limited by many factors, such as the harsh environmental requirements, very low load capacity, and difficulty in manufacturing ideal surfaces of large area. Therefore, research in the near future should focus on improving the superlubricity properties of solid lubricants.

In contrast to solid superlubricity, liquid superlubricity has greater application potentials in mechanical systems, because it is relatively insensitive to environment and has a higher load capacity com-

pared to the solid variety. However, the mechanism of liquid superlubricity is still imperfectly understood, with many problems to be solved. At the nano-scale, hydration lubrication has been proven one of the most effective ways to achieve liquid superlubricity. The main mechanism is attributed to the repulsive hydration force and the formation of a fluid hydration layer. However, this mechanism is unsuitable for macroscale superlubricity. Instead, Luo et al. found that the formation of hydrogen-bond networks is also a reasonable explanation for macroscale liquid superlubricity. One typical representative system is phosphoric acid solution. Of course, the model of hydrogen-bond networks cannot cover all liquid superlubricity phenomena at macro-scale. For example, the superlubricity mechanism of ceramics with water lubrication is mainly attributed to the formation of silica layers and hydrodynamic lubrication. In fact, recent work by Li et al. showed that pure hydrodynamic lubrication could also achieve superlubricity under certain conditions, independent of lubricant type. The mechanism underlying liquid superlubricity is much more complicated than that of solid superlubricity, and thus deserving of closer study in the future. The final aim of liquid superlubricity is to design promising mechanical systems with super-low friction and wear, but the application of superlubricity to mechanical systems remains challenging, as noted by Prof. Adachi. Therefore, new technologies applying liquid superlubricity to various mechanical systems may become a common and important research topic in the future.

In summary, this workshop provided both a platform for the exchange of academic achievements between international scholars in tribology and a vital chance to exhibit research progress in various branches of science. This workshop is significant in accelerating the development of superlubricity.

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