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Observation of the Amontons-Coulomb law on the nanoscale: Frictional forces between MoS₂ flakes and MoS₂ surfaces

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Abstract. – The MoS₂ flake moves on MoS₂ in such a way that the stacking of the sulphur basal plane of MoS₂ is maintained. The frictional forces between MoS₂ surfaces are clearly proportional to the loading force although they depend strongly on the movement of the MoS₂ flake. The Amontons-Coulomb law is excellently satisfied at loading forces in the range of 1–120 nanonewtons. The frictional coefficient between MoS₂ surfaces along the direction $[10\bar{1}0]$ of the MoS₂(0001) surface is estimated to be approximately 0.003.

It has long been known that the layer materials such as graphite, muscovite mica and MoS₂ exhibit small friction. However, the reason why they exhibit small friction has not been clarified in full because the friction occurring at layer materials involves grains in the interface and surface deformations. In the case of MoS₂, with the exception of a paper [1] indicating that ultralow friction occurs at MoS₂ flakes on MoS₂, there are few experimental studies on friction between MoS₂ surfaces despite the fact that MoS₂ is a very important lubricant. In this paper [1], it has been reported that ultralow friction with a friction coefficient of 10^{-3} order is caused by the friction-induced orientation of easy shear basal planes of the MoS₂ crystal structure parallel to the sliding direction.

In this letter, we report that a MoS₂ flake moves on MoS₂ in such a way that the stacking between sulphur layers of MoS₂ is maintained. These frictional-force maps from the MoS₂ flake on MoS₂ (surface friction) exhibit a pattern quite different from that of a tip on MoS₂ [2] (henceforth called tip friction). We here demonstrate, for the first time, that the Amontons-Coulomb law is excellently satisfied at loading forces in the range of 1–120 nanonewtons, based on the atomic-scale movement of a MoS₂ flake.

A MoS₂ substrate was prepared by cleaving a natural MoS₂ block. A MoS₂ flake with an area of 1mm square and a thickness of several micrometers was used. Normal and lateral forces were measured simultaneously under a relative humidity of below 50% at room temperature, using a commercially available instrument (Seiko Instruments Inc., SPI-3700). There was not a prominent difference between the lateral forces measured under different relative humidities. The scan speed was 0.13 $\mu\text{m/s}$. The lattice orientations of the upper MoS₂ flake and lower MoS₂ substrate were not specified with respect to the sliding direction because the scan direction for a MoS₂ flake on the MoS₂ substrate was arbitrarily taken. A rectangular silicon cantilever with a Si₃N₄ tip (a normal spring constant: 0.75 N/m) was used. Zero normal

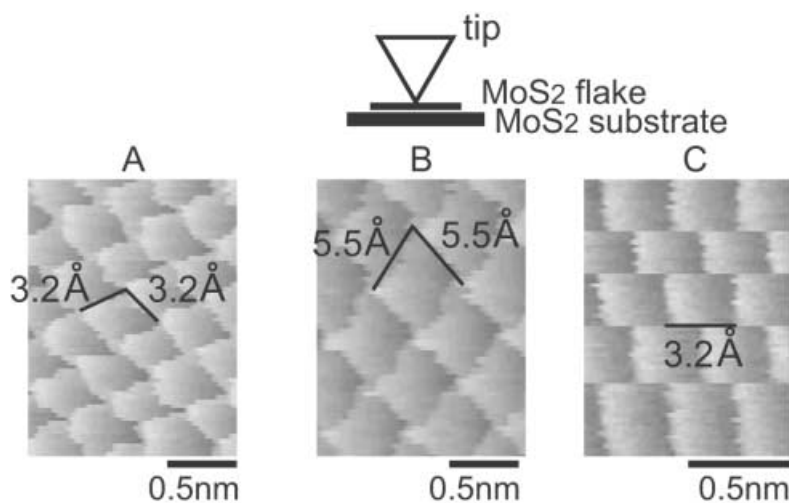


Fig. 1 – Three x frictional-force maps at different areas from a MoS₂ flake on MoS₂ obtained by scanning along the x -direction.

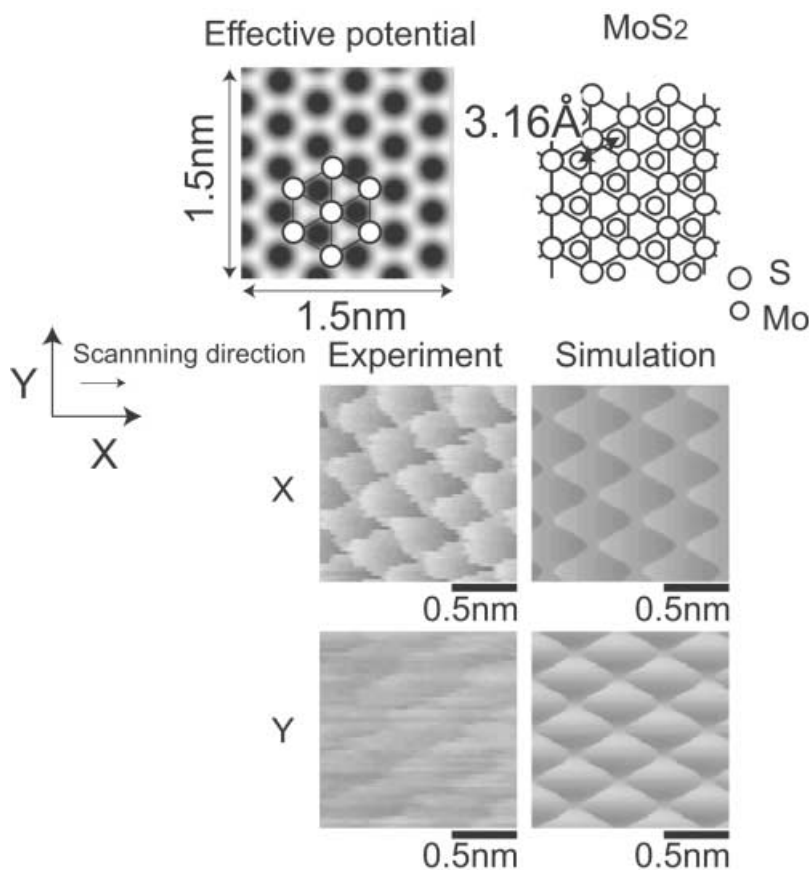


Fig. 2 – Experimental data and theoretical simulation for type A of fig. 1.

force is defined as the position at which the cantilever is not bent. The frictional forces were calibrated by using the method of Meyer *et al.* [3].

Figure 1 shows three different x frictional-force maps from a MoS₂ flake on MoS₂ scanned along the x -direction. It should be noted that these exhibit scaled (types A and B) and square (type C) patterns, clearly different from that of a tip on MoS₂ from the standpoint of a periodic pattern and contrast (see figures on the left-hand side of fig. 4 below). Noting that this system consists of friction between a tip and a MoS₂ flake and that between a MoS₂ flake and MoS₂, it was found that the friction from this system includes two different mechanisms. However, it can be concluded that the frictional features of a flake on MoS₂ exhibit only friction between a MoS₂ flake and MoS₂ because they do not include the frictional-force map from a tip on MoS₂. This means that the friction between a tip and a MoS₂ flake is greater than that between a MoS₂ flake and MoS₂. Now, we consider type A of the frictional-force map, which is the standard pattern appearing in this experiment. The movement of the flake on MoS₂ is substituted into the movement of a single particle moving with a constant velocity in the effective potential for type A, as follows: $V = V_0 \left\{ 2 \cos \left(\frac{2\pi}{a} x \right) \cos \left(\frac{2\pi}{a\sqrt{3}} y \right) + \cos \left(\frac{4\pi}{a\sqrt{3}} y \right) \right\}$,

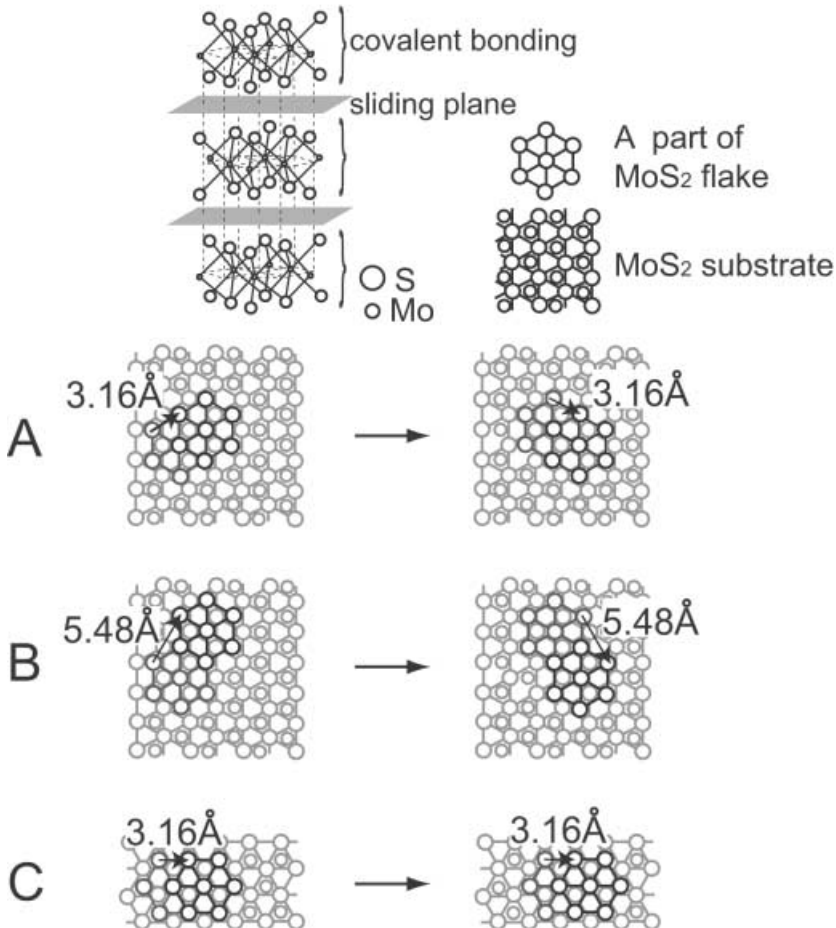


Fig. 3 – The three types (A, B and C) of the MoS₂ flake movement are shown; the arrows represent the scanning direction.

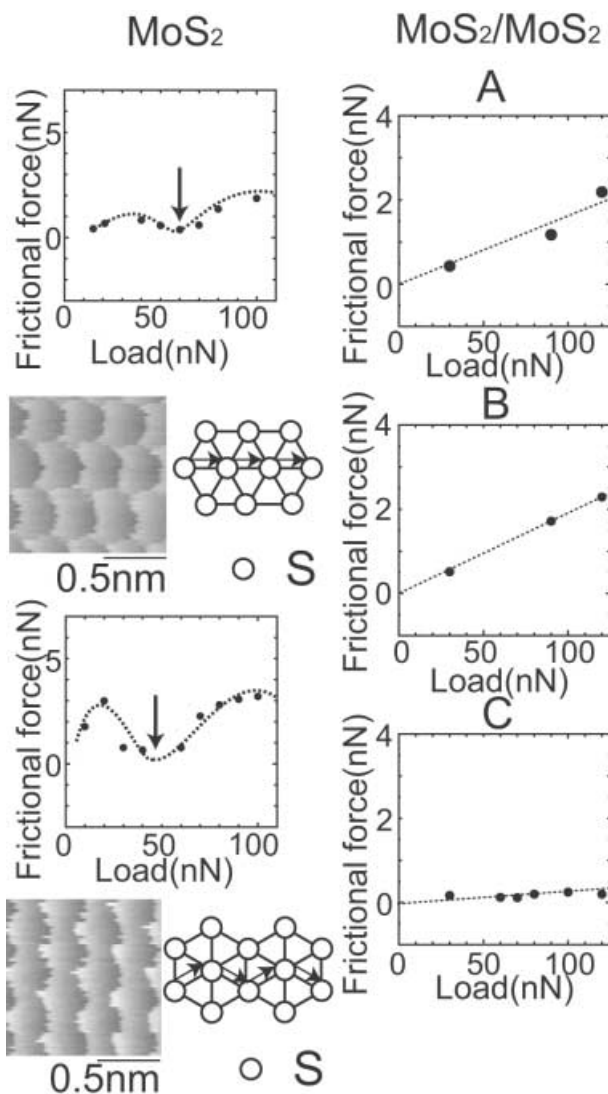


Fig. 4 – The frictional forces in relation to the loading force for the surface friction (right-hand side) and for the tip friction (left-hand side). Images and arrows on the left-hand side represent frictional-force maps and the tip movement, respectively.

where $a = 0.316$ nm. This effective potential is determined so that it takes the minimum values at the natural stacking relation of MoS_2 . The result of this simple preliminary simulation is in excellent agreement with the experimental data shown in fig. 2. Here, the spacing of $a = 0.316$ nm indicates a jump to the next stable position maintaining the stacking of MoS_2 , as shown in fig. 3. Similarly, it is also revealed that the movements for types B and C are performed in such a way that the natural stacking relation of MoS_2 is maintained, as shown in fig. 3.

The mean frictional forces for different movements in relation to the loading force are shown on the right-hand side in fig. 4. These are all proportional to the loading force although they depend strongly on the movement of the MoS_2 flake. Thus, the proportional coefficients

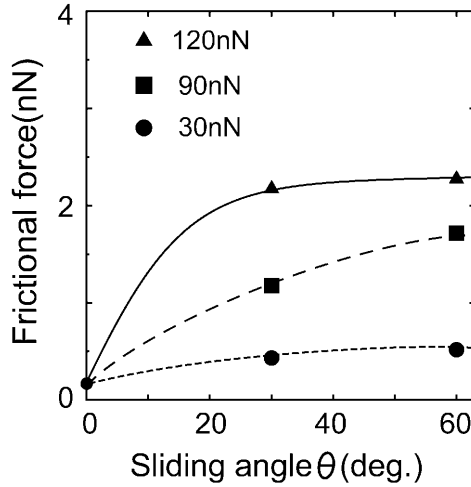


Fig. 5 – The frictional forces *vs.* the sliding angle deduced from the data of fig. 4, where the sliding angles for cases A, B and C are 30° , 60° and 0° , respectively.

defined as μ_A , μ_B and μ_C for the cases A, B and C are estimated to be approximately 0.01, 0.02 and 0.003, respectively. The proportional coefficient μ_C for the case C is much smaller than μ_A and μ_B for the cases A and B, which corresponds to the frictional coefficient $\mu_{10\bar{1}0}$ and is close to the experimental value (below 0.002) obtained by Martin *et al.* [1]. Thus, it is found that the $\text{MoS}_2(0001)$ flake is easy to shear along the direction $[10\bar{1}0]$ of the $\text{MoS}_2(0001)$ surface. Surprisingly, the frictional coefficient for the case B is eight times larger than that for the case C. Proportionality of the frictional forces in relation to the loading force also reveals that the contact area between MoS_2 surfaces S is proportional to the loading force F_N , using the relationship $F = \mu F_N = \tau S = \tau \alpha F_N$, where α represents the proportional constant. This indicates that surface deformation is significantly smaller in MoS_2 than in graphite because the lamellar MoS_2 consists of covalent bonding, which behaves like a hard plate. Along the different scanning directions of the $\text{MoS}_2(0001)$ surface, a tip performs one-dimensional stick-slip and zigzag stick-slip motions, shown on the left-hand side in fig. 4. This behavior is similar to that of ionic surfaces [4]. In the case of an ionic surface, the minimum points (shown by the arrows) appear before large distortions of the surface occur. Thus, it is inferred that large distortions of the surface occur even in MoS_2 triple layers after the minimum points shown by the arrows. Furthermore, it is interesting to note that these results are in good agreement with a recent computer simulation result by Wenning *et al.* [5], *i.e.*, shear forces for tip friction have a strongly oscillatory component and shear forces for surface friction obey the Amontons-Coulomb law of friction.

Now, the anisotropy of the frictional force for a different directional movement of the flake *vs.* the pulling direction is shown in fig. 5 from the results of fig. 4, which appears depending on the sliding angle θ , denoted as the angle between the direction of the flake movement and the pulling direction. One notes that the frictional forces *vs.* the sliding angle increase strongly with a higher load. Furthermore, the frictional forces are found to increase with the increase of the sliding angle, which, interestingly, is consistent with the frictional mechanism between flat surfaces discussed by Gyalog and Thomas [6]. These results lead to the following two points: i) in the case where the flake movement is parallel to the pulling direction, the frictional force is smallest. ii) In the case where the direction of the flake movement is not parallel to that of the pulling force, the flake does not move toward the next stable point

until the projection of the pulling force is coincident with the force necessary to begin moving toward the next stable point. Thus, the larger the sliding angle, the larger the pulling force. In case B, the energetic barrier is supposed to be very large because the path to the next stable point takes the path of the stacking (S atom above S atom). Here, it may make sense to discuss the term “incommensurability” introduced by Hirano *et al.* [7]. They measured the frictional forces of muscovite mica as a function of the lattice misfit angle between the two contacting cleavage lattices and found that the frictional forces are anisotropic with respect to the lattice misfit angle, *i.e.* the frictional forces increase (decrease) when the surfaces approach being commensurate (incommensurate). However, the appearance of the anisotropy of the frictional force in this experiment reveals that a MoS₂ flake on MoS₂ always takes the initial and final positions so that the stacking of the sulphur basal plane of MoS₂ is maintained. Thus, the two contacting MoS₂ surfaces are always commensurate and the sliding direction of the MoS₂ flake does not always coincide with that of the pulling direction, which may be different from the conclusions of Hirano *et al.*

Furthermore, the results obtained here may give an explanation to the problem why the friction of the system including various flakes is generally small, *e.g.*, a friction force acting on many flakes with different movements may be close to zero, which is consistent with a friction coefficient of 0.001 for two MoS₂ surfaces with poly-crystalline microstructures obtained by Le Mogne *et al.* [8].

This study reveals the change in the frictional behavior when the frictional mechanism is transient from a single-atomic tip to a large-area flake tip (compare data on the left-hand side in fig. 4 with those on the right-hand side). There is a prominent difference between frictional-force maps of tip friction and those of surface friction. In this study, we did not treat the flake movement as that of an elastic material but as that of a rigid material; nevertheless, we could understand the movement successfully. The MoS₂ flake moves on MoS₂ so that the stacking of the sulphur basal plane of MoS₂ is maintained. However, the frictional forces between MoS₂ surfaces are proportional to the loading force and depend strongly on the movement of a MoS₂ flake. The frictional coefficient $\mu_{10\bar{1}0}$ at the sliding angle $\theta = 0^\circ$ is estimated to be approximately 0.003, which indicates that a MoS₂ flake is easy to shear along the direction $[10\bar{1}0]$ of the MoS₂(0001) surface. These findings also provide us important information of how to decrease friction between MoS₂ surfaces.

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