Research on the origin of solid super-slip and friction based on Raman spectroscopy

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Abstract. It is estimated that about 1 / 3 ~ 1 / 2 of the world's energy consumption is caused by friction and wear. About 80% of the parts of general machinery and equipment are scrapped due to wear. In China, the annual loss caused by friction is more than 4 trillion. Therefore, to explore the origin of friction and realize solid super sliding has become a key point of people's attention. But the basic scientific principle and mechanism of friction origin are difficult to explore, so how to give a reasonable method to solve this problem is particularly important. This paper focuses on the use of Raman spectroscopy to observe the solid super slip phenomenon and phonon dissipation, through Raman spectroscopy to determine the orientation of molecules on the friction interface, detect the related properties of two-dimensional materials, as well as crystal defects and their apparent morphology to explore the origin of friction, and to reduce the energy loss caused by friction.

1. Introduction
The origin of friction and its influencing factors are fundamental issues in tribological research. In recent years, the frictional energy dissipation model used to understand the origin of friction has gradually become the consensus of researchers. This model focuses on the dissipation of various frictional energy such as phonons and electrons. It is considered that phonon dissipation is the most important process among them. In order to study this process, it is not enough to be limited to theoretical simulations. Corresponding experimental methods and means must be available. On the other hand, the solid super-slip system with two-dimensional materials as the main focus has contributed a lot of valuable phenomena and mechanisms to tribology, but there are more hotspots to be developed for two-dimensional material systems. The understanding of the influence of the related properties of two-dimensional materials on friction also requires effective methods for testing various properties of two-dimensional materials. Raman spectroscopy is an experimental method to meet these needs.

2. Solid super slip and the origin of friction
The concept of solid super-slip was first proposed by Hirano and Shinjo in the early 1990s. Based on the Frenkel-Kontorova-Tomlin-son model, they theoretically predicted when two crystal planes move relative to each other in certain directions. The crystal planes are in a state of incommensurability, the friction force will completely disappear, and this state is called superlubricity [1]. This theory also caused a great sensation among scholars at that time, because Once a method of overcoming friction is discovered, the whole world will benefit a lot. However, absolute zero friction is impossible in reality. However, if the principle of ultra-slip is used, the friction will be reduced by orders of magnitude. It is also an extremely important contribution. After a series of theoretical investigations and experimental
material explorations, people first discovered super-slip properties on two-dimensional materials, such as molybdenum disulfide and graphene. The reason why two-dimensional materials exhibit super-slip phenomenon can be realized because the interaction force between the two-dimensional material layers is very weak and incommensurate contact can be achieved. Therefore, the structural factor of incommensurability is considered to be one of the main mechanisms of solid super-slip. It has also been found that this structural factor is very important for the friction performance between the layered structure or the single crystal interface, so the academic circles call this type of super-slip structure super-slip. The earliest investigation of solid super-slip phenomenon can be traced back to 1993, when Martin et al. [2] first experimentally verified the existence of solid super-slip. They achieved ultra-low friction of the order of 0.001 with MoS2 under ultra-high vacuum. Coefficient, they attribute it to the anisotropy existing on the sulfur-rich base surface. Subsequently, Dienwiebel et al. [3] used an atomic force microscope as a tool to achieve super-slip in graphite, and proved that this super-slip is anisotropic to the scanning direction.

Where the friction force originated is a question that people have always been concerned about. In 1699, the French physicist Amontons made a very influential report. He proposed that the friction force comes from the positive pressure it receives and the size of the object. It is irrelevant, and it is considered that the two micro-protrusions on the uneven surface mesh with each other, and the relative movement must raise the load from one meshing position to another meshing position, thereby causing frictional energy loss [4]. It was an inspiration for people, and then many famous theories were put forward, such as mechanical meshing theory, adhesion theory, etc. The friction energy dissipation is a theoretical model for understanding atomic-scale friction phenomena proposed in this context. This model believes that the friction process is nothing more than a process in which the kinetic energy (or mechanical energy) of a moving object is transformed into other forms of energy. The medium of this process is various types of "primitive excitations" induced by mechanical excitation, such as phonons, electrons, and photons. As well as excitons. They can generate local vibrations of surface atoms or molecules adsorbed on the surface, and can further cause various processes such as the break of chemical bonds on the surface, the reconstruction of molecular orientation, and the occurrence of fluorescence. Almost all energy consumption The dispersion path is related to the heat energy carried by various vibrations at the atomic scale. The behavior of these vibrations or heat transfer, including its propagation characteristics and energy characteristics, needs to be described by the concept of phonons, so the characteristics of phonons are understood. Including its population rule and dynamic process under specific excitation, it is very important to understand the process of friction energy dissipation. Phonon dissipation is the main way of friction energy dissipation.

In 1929, Tomlinson [5] proposed the independent vibrator model, which is often used to simulate and calculate the atomic-scale friction phenomenon, which is the most concerned model. He regarded the interaction on the friction interface as a spring connected to a rigid support. The interaction between the atoms and the periodic potential field, and when these atoms move in the periodic potential field, the transformation from spring deformation energy to atomic vibration energy occurs continuously, so the excitation of vibration makes energy irreversibly dissipated in the solid in the form of phonons Dispersed. The independent oscillator model simply thinks that the vibrations of the atoms on the interface do not affect each other and proceed independently. Based on this model, the composite oscillator model developed by Frenkel and Kontorova [6-7] not only considers the atoms It also replaces the periodic potential field in the model with a series of interface atoms connected by a spring network. Although the composite vibrator model is more complex in form, it does not change the core feature of the transformation of spring deformation energy to vibration energy, and also believes that phonons are the main way of energy dissipation.

3. The basic principle of Raman scattering and its application in the study of the origin of friction

Raman scattering is a relatively common method to detect vibration and phonons. The basic principle is as follows. When a beam of light enters a substance, it usually scatters, that is, it deviates from the original direction and continues to propagate. When the wavelength of the scattered light is the same as
the wavelength of the incident light, and the energy of the incident photon does not change after interacting with the substance, the scattering is called elastic scattering. In contrast, if there are a few photons and the molecules (atoms) or solids in the object When the energy exchange occurs in the lattice, the frequency of the emitted photons will change. This phenomenon is called inelastic scattering, and because this phenomenon was first discovered by Indian physicist Raman, it is also called it Raman scattering. When the energy exchanges between matter and incident light, roughly two situations occur. The first situation is to absorb energy to the incident light, and the other is to transfer energy to the incident light. Therefore, according to these two situations, we can use Raman Scattering is further subdivided. When the energy is transferred from the incident light to the material, the energy of the incident photon decreases, which will cause the frequency of the scattered light to decrease. We call this part of the scattering as Stocks scattering. When the energy is transferred from the material to the incident light, Increasing the energy of incident photons leads to an increase in the frequency of the scattering spectrum and a decrease in wavelength, which we call Anti-Stocks scattering as shown in Figure 1.

However, because the probability of Raman scattering is very weak, we need some lasers with high power density to act, so that it can be fully amplified before it can be applied to the detection of matter. From the perspective of quantum mechanics to analyze Raman scattering, its occurrence What time interacts with photons is actually the vibrational energy level of matter. This process can be regarded as the effect of photon, the matter transitions from an unstable virtual energy level among different energy levels, and Stocks scattering produces The final effect of is that matter transitions from a lower energy level to a higher energy level. Anti-Stocks is the opposite. Matter transitions from a higher energy level to a lower energy level. And because the matter is in The probability of being distributed at higher energy levels is very low, so the probability of occurrence of Anti-Stocks is much weaker than the probability of occurrence of Stocks. Phonons are used as a measure of photons propagating in the crystal, and the behavior of phonons is represented by the frequency of wave deviation. In crystals, the phonon spectrum is generally drawn in the direction of high symmetry according to the dispersion relationship. The usual
Raman spectroscopy measurement requires the use of visible light to excite the sample. The wave number of the vibrating phonon represented by the conservation of momentum and excitation The wave number of light belongs to the same order of magnitude, about $10^{-5}$ cm$^{-1}$. The phonons that can be measured by Raman spectroscopy belong to the phonons under the wavelength limit, and only occupy a small part of the Brillouin zone, that is, Raman spectroscopy measurement Is usually the nature of phonons with wave loss distributed near the center of the Brillouin zone.

3.1. Determine the molecular orientation on the friction interface

The scope of application of two-dimensional materials is not only included in the solid field, and its extraordinary characteristics in the field of liquid lubrication are also very superior. For example, adding molybdenum disulfide nanosheets as a lubricant additive to the base oil polyalphaolefin-8 (PAO-8), and testing the effect of its addition on the tribological properties of lubricating oil, the results show that for molybdenum disulfide nanosheets, when the addition amount is 0.20mg/ml, the wear rate of lubricating oil is reduced to 50% of the original pure base oil. The mechanism of action is: under boundary lubrication or thin-film lubrication conditions, lubricating oil molecules will be physically or chemically adsorbed to form a boundary film on the friction surface, blocking the direct contact between the two friction pairs, thereby acting Anti-wear effect. And because the molecules in the boundary film tend to adopt the same orientation perpendicular to the friction surface, and they will tilt and change their orientation during the sliding process, the measurement of the molecular orientation in the actual lubricating interface is of great significance for understanding the mechanism of lubrication. So how to measure the molecular orientation in the lubricating interface? After years of discussion and research, scholars have discovered that because the signal intensity of the Raman spectrum is directly related to the polarization direction of the excitation light and signal light, the molecular orientation can be measured by measuring the change of the Raman peak of the crystal vibration, such as: BA Kolesov [8] measured the orientation of the CuPc film deposited on the silicon substrate by measuring the variation of the Raman peak corresponding to the vibration of the β-CuPc crystal under different polarization directions. However, because the lattice structure of some molecules is not clear when they form a crystal, they soon improved the method. By measuring the incident and scattered light polarization directions are parallel to each other and perpendicular to each other, the Raman corresponding to the internal vibration mode of the molecule The ratio of the intensities of the peaks gives the molecular orientation of the two configurations of α and β CuPc [9]. After them, Shan Jiang [10] et al. used a polarized confocal Raman system to measure the orientation of PDMS chains when adsorbed on the (001) crystal surface of mica, and found that PDMS molecules are not completely disorderly arranged on the mica surface, but To a certain extent, there is a more obvious orientation tendency, which leads to the obvious anisotropy of the Raman signal given by PDMS on the mica surface. After years of research and improvement, Tsinghua University scholar Zhang S[11] and others combined the Raman spectroscopy system with the traditional ball/disk paired grinding device to realize the liquid crystal molecules (5CB) that act as lubrication in the friction contact area. For real-time detection of orientation, the experimental system is shown in Figure 2.
Figure 2  (a) schematic diagram of real-time Raman detection system for molecular orientation of lubricating liquid film; (b) 6CB adsorbed silver nanorods; (c) Diagram of lubricant film delamination in contact area in film lubrication model

When the relative movement speed between the balls and the disk exceeds a certain threshold, the liquid crystal molecules in the contact area obviously show a tendency to align in the direction of rotation, and the higher the rotation speed, the more obvious the orientation. On the periphery of the contact area, the liquid crystal molecules maintain a disorderly arrangement.

3.2. Testing the relevant properties of two-dimensional materials
Measurements with the help of low-wavenumber Raman spectroscopy can provide information about the interaction between two-dimensional materials, because the interaction between layers can induce characteristic vibrations in two-dimensional materials. However, the frequency of the vibration caused by van der Waals force must be low, so the difficulty in measuring this low-frequency vibration lies in how to use optical means to distinguish the scattered light signal from the Rayleigh scattering which is dominant in the scattered light. The main methods currently used are to use a spectrometer with a relatively long focal length for more effective spectroscopy [12], or use a multi-stage notch filter with an ordinary spectrometer to minimize the influence of the excitation light on the signal [13]. The measured low-wavenumber Raman signals of two-dimensional materials can be divided into shear mode (all molecular layers vibrate in the horizontal direction) and breathing mode (all molecular layers vibrate in the vertical direction) according to the vibration direction of each molecular layer. The wave numbers of both vary with the number of layers of the two-dimensional material, as shown in Figure 3.

Figure 3 (a) S-shear mode, B-breath mode, TL-trilayer of low wavenumber Raman spectra of WSe2, which means that each layer of WSe2 contains three atomic sublayers; (b) Linear atomic chain models of 2-layer and 3-layer MoS2
There is a quantitative relationship between the wave number and the number of layers of two-dimensional materials. This quantitative relationship can be derived from a relatively simple semi-classical theoretical model, that is, the linear atomic chain model. This relatively simple model is actually a more general crystal. Simplification of lattice dynamics model [14]. When Zhang Xin [15] et al. used these two methods at the same time to analyze the low wave number mode of MoS2, they found that the two methods gave the same results. This reflects the fact that the low-wave number vibration originates from the interlayer interaction (namely, Van der Waals force) and has little to do with the covalent bond in the layer. Based on this characteristic, the second method is basically adopted in a system containing multiple atomic sublayers in a single layer, which is easier to solve. In most two-dimensional material systems, the force constant characterizing the interaction between layers is on the order of $10^{19}$ N/m$^3$.

3.3. Crystal defects and inspection surface morphology

Crystals will inevitably produce a certain number of defects during the nucleation, growth process, or use of materials, which will affect their physical properties and material properties. To be more specific, defects in the crystal material will cause local deformation and stress. This has an impact on the mechanical properties of materials, transportation behavior and other physical properties, which is particularly significant in low-dimensional materials. The two-dimensional material is a low-dimensional structure that has attracted much attention in recent years. Therefore, studying the defects of two-dimensional materials in various situations is an essential step. First, LG Cancado [16] and others bombarded graphene with different concentrations of argon ions (Ar+) to produce different numbers of defects in the graphene, and used Raman spectroscopy to characterize these graphenes and found that the obtained Raman spectra The ratio of the intensity of the D peak and the G peak is related to the defect density in graphene and the wavelength of the excitation light. Based on the experimental results, they also gave the quantitative satisfaction of the peak intensity ratio, excitation light photon energy and point defect density. The expression is shown in Figure 4.

Figure 4 Raman spectroscopy is used to detect defects and stresses. (a) Differences in Raman spectroscopic morphology of graphene under different point defect densities; (b) The relationship between point defect density, Raman peak intensity ratio and Raman excited photon energy; (c) Relationship between graphene characteristic peak position and biaxial stress; (d) Bubble loading and stress characterization device schematic.
Later, A. Eckman [17] et al. experimentally studied that the type of defects in graphene also affects its Raman spectral characteristics. Here, the intensity ratio between the D peak and the D' peak is used to characterize the defect type: for sp³ defects (i.e., due to fluorination or oxidation, the original sp² hybrid orbitals of some carbon atoms in the graphene molecular layer are changed to sp³ hybrid orbitals. The ratio is the largest, which is 13; for hole defects, the ratio is 7; this peak intensity ratio is the smallest in grain boundary defects, which is 3.5. In terms of stress measurement, J. Zabel [18] et al. conducted a systematic measurement of graphene bubbling on a silicon oxide substrate, and found that applying isotropic biaxial stress to graphene will cause the G peak in the Raman spectrum and the position of the 2D peak is red-shifted, and the offset is linearly related to the magnitude of the applied stress. After them, G. Wang [19] and others developed a sealed bubble loading device based on graphene covering micro pores. At the same time, the results of AFM topography scanning and the change of the G peak of the graphene Raman spectrum reflect the graphite stress in the alkene realizes the characterization of the interlayer shear stress of the double-layer graphene. Based on the summary and enlightenment of predecessors, S. Zhang [20] and others went a step further and measured the friction force of bubble graphene that was loaded with different stresses to produce different degrees of strain, and found that the friction force and friction coefficient increase with increasing strain means that the super-slip state of graphene can be adjusted through the control of strain.

In the research on using surface texture and other surface technologies to reduce friction and wear, effective methods are needed to characterize the morphology of the prepared surface structure. The surface morphology not only directly affects the performance of the material, but also affects the quality, reliability and life of the material. Therefore, how to accurately and reliably detect the morphology has become an urgent problem to be solved. After a large number of experiments, Motohiro Banno [21] et al. combined the two methods of stimulated Raman and optical interference detection, and developed a differential coherent contrast stimulated Raman microscope (DIC-SRS), as shown in Figure 5.

![Figure 5 DIC-SRS is used for surface morphology detection. (a) schematic diagram of measurement principle; (b) Detection results of Si surface with grooves](image)

The device coherently superimposes the stimulated Raman signal at the focus of the objective lens with a beam of reference light. The intensity of the superimposed signal is different at different positions.
on the surface due to the difference in the path of the light. They successfully combined this method applied to the detection of Si surface structure, a high resolution of about tens of nanometers is obtained. In addition to the higher vertical resolution, this method can also be used to detect the morphology of the interface buried under the liquid film.

3.4. Characterization of phonon dynamics

The dynamic process of phonons is mainly the relaxation behavior of phonons. After a lot of research, it is found that relaxation has an important influence on the dissipation of frictional energy. However, the general method alone cannot accurately measure the relaxation behavior of phonons. In the end, scholars learned through the elimination method that Raman measurement has a chance. This method was first used by JA Kash [22] to measure the relaxation behavior of phonons in GaAs with a picosecond time-resolved Raman technique, and proposed that the avalanche process of phonon population controls the energy relaxation in GaAs. In the Henan process, phonons are the main way electrons transfer energy to the lattice. He found that within a certain range, the population of non-equilibrium phonons maintain a linear relationship with the carrier concentration generated by light excitation. They also studied the relationship between the phonon relaxation process and temperature. They measured the attenuation curves of phonons at room temperature and at a low temperature of 20 K refrigerated by liquid nitrogen, and found that the relaxation times of phonons in the two cases were 4.0 ps and respectively. 7.5 ps. As shown in Figure 6, it is verified that the decay rate of the non-equilibrium phonon population decreases with the decrease of temperature.

![Figure 6](image)

The phonon relaxation process. (a) The lifetime of LO phonons in GaAs at room temperature and low temperature; (b) The change of population and vibration frequency of phonons of graphite G mode with time

Then K. T. Tsen [23] et al. gave similar conclusions on GaAs quantum wells. They measured the relaxation time of phonons at a low temperature of 10 K to be about 8 ps. H. Yan [24] et al. used time-resolved Raman spectroscopy to study the dynamic behavior of graphene phonons. They measured that the phonons were established within about 300 fs after being excited. It is in a highly unbalanced state and will decay with a relaxation time of about 2.2 ps; at the same time, the vibration frequency also decreases with time, as shown in the figure. In such a short relaxation time, due to the weak interaction between the graphite layers, the energy transfer process occurring between the layers is almost negligible. The difference is that C. Ferrante [25] and others used the FSRS system to study the energy relaxation process of hemoglobin in biological macromolecule systems, and proposed that in biological molecules such as hemoglobin, the energy obtained by excitation light is also through specific The redistribution occurs due to the relaxation of the vibration mode population, which provides a new perspective for understanding the energy redistribution process of biomolecules.

With the continuous deepening of the experiment, Chinese scholars also have their own unique discoveries, such as S. Wu [26] and others through the Raman spectrum measurement carried out in graphene. They found that for phonons, the phonon relaxation time will be shortened as the number of graphene layers decreases; they also found that compared to graphene directly attached to the surface of SiO2/Si, the same number of graphite layers suspended on the hole The alkene sample has a longer phonon relaxation time, as shown in Figure 7. Their explanation is that the attached graphene can convert
the energy carried by the phonons through the additional channel of the graphene-SiO2 interface, so the phonons will relax faster.

Figure 7  (a) Time-resolved anti-Stokes Raman spectra of graphene; (b) The phonon lifetime difference in different layers of graphene; (c) Effect of substrate on phonon lifetime; (d) Effect of doping and defect on phonon lifetime

4. Conclusion

The origin of friction and its influencing factors are fundamental problems in tribology research. In recent years, the friction energy dissipation model used to understand the origin of friction has gradually become the consensus of researchers. The model focuses on phonons, electrons and other ways of friction energy dissipation, and considers that phonon dissipation is one of the most important processes. In order to study this process, it is not enough to limit theoretical simulation, and there must be corresponding experimental methods and means. On the other hand, solid super-sliding systems with two-dimensional materials as the main object of concern have contributed a lot of valuable phenomena and mechanisms for tribology. However, two-dimensional material systems still have more hot spots to be developed. In order to expand the current understanding of the impact of two-dimensional materials on friction, it is also necessary to have effective methods to test various properties of two-dimensional materials. Raman spectroscopy is an experimental means to meet these needs.

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