Ripple formation induced in localized abrasion

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The formation of nanometer-scale patterns while scratching a KBr(001) surface with a scanning force microscope in ultrahigh vacuum is reported. Wear of single atomic layers has been observed when the microscope tip is repeatedly scanned across a line. The initially flat surface is rearranged in a quasiperiodic pattern of mounds and pits. The distance between the pits is about 40 nm when normal forces of a few nanonewtons are applied, and it slowly increases with the load. If a square area is scanned, a pattern of ripples is formed. These features can be interpreted within an erosion process induced by a periodic increase of the strain produced by the scanning tip.

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I. INTRODUCTION

Ripple formation due to perturbations acting on a surface is commonly observed on macroscopic scales. Well-known examples are given by wind-blown sand dunes observed in the desert and on the shore.[1] The dynamics of sand motion consist of two processes: the transfer of the sand grains from one position to another, and the movement of the sand along the surface without jumps. The combination of both results in the formation of regular dunes. Elastic instability waves were observed by Schallamach on macroscopic length scales for elastically soft materials such as rubber during sliding on hard surfaces.[2] Self-organization of surface undulations have also been reported on the nanometer scale. For example, ripples are formed when glasses, amorphous films, semiconductors, or metals are sputtered by ion beams.[3] These features have typical wavelengths of few tens of nanometers, and they can be revealed by scanning probe microscopes.

Leung and Goh observed the formation of ripples when the tip of an atomic force microscope (AFM) was scanned over a polymer film.[4] The orientation of the ripples was perpendicular to the scan direction and their characteristic wavelengths were in the range between 10 and 100 nm. Ripples produced by scratching were observed on polymers several times, and, more recently, even on gold films.[5] The ripples were considered to be the result of a peeling process operated by the microscope tip in the case of polymer films,[6] or as a result of a self-regulating periodic pickup and release of clusters in the case of gold films.[5]

II. EXPERIMENTAL RESULTS

Here, we report the formation of ripples across scratches on ionic crystal surfaces. Scratches were produced and imaged using a home-built force microscope in ultrahigh vacuum (UHV) at room temperature.[7] We used commercially available silicon cantilevers with normal and torsional spring constants of about $c_n = 0.09$ N/m and $c_t = 49$ N/m. The radius of curvature of the tips was nominally below 15 nm (Nanosensors). The same cantilevers were used for both scratching and imaging the modified areas. Two different KBr(001) surfaces were studied. Sample A was cleaved *in situ*, while sample B was cleaved in air and immediately transferred into the UHV chamber. Both samples were heated to 120 °C for 15 minutes. The different cleavage procedure affected the morphology of the surface significantly; on the sample cleaved in UHV atomically flat areas not wider than 100 nm were observed, whereas the surfaces cleaved in air revealed flat terraces larger than 1 μm. However, atomically clean surfaces are obtained by both methods.[8]

Figure 1(a) shows the topography image of a groove on the surface of KBr(100), recorded after scratching the surface 512 times with a normal load $F_N = 26.6$ nN and a scan

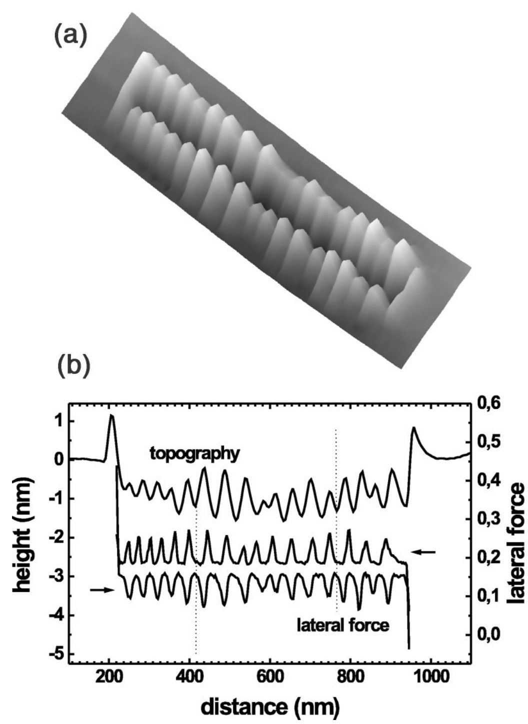

FIG. 1. (a) Topography image of a groove formed on KBr(001) after 512 scratches along the (100) direction with $F_N = 26.6$ nN and $v = 2.67$ μm/s (sample A). (b) Cross section of the topography along the ground of the groove and lateral force acquired while scratching the groove. The lower and upper parts of the lateral force loop were obtained while scanning forwards and backwards, as indicated by the arrows. The dotted lines indicate positions in the pits where the tip is decelerated as derived from an increase of the lateral force in either direction.

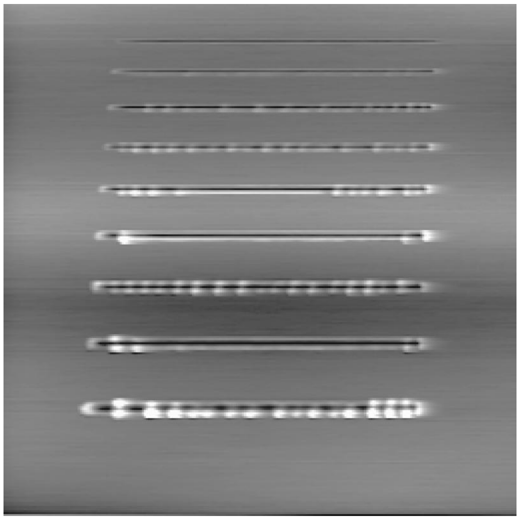

FIG. 2. Topography map of a region scratched with the following loads (up to down): 1.7, 5.3, 8.8, 12.4, 15.9, 19.5, 23, 26.6, 30.1 nN. Frame size: 1.2 μm.

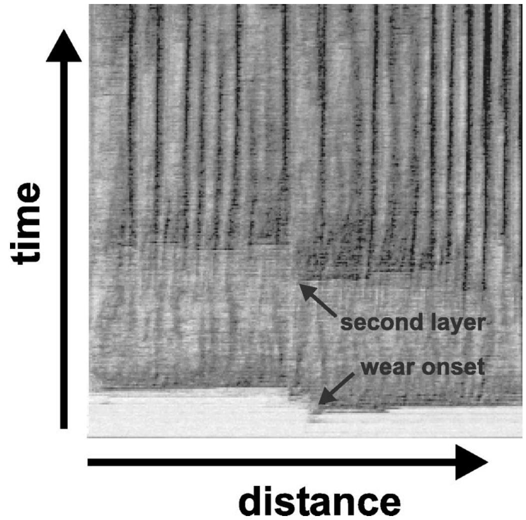

FIG. 3. Development of lateral force with time while scratching a single line of 800 nm length forwards and backwards. In horizontal direction the lateral force during forward scan is plotted, where darker color means stronger lateral force; the vertical direction is the time axis.

velocity $v=2.67$ μm/s. The groove is surrounded by a pattern of 34 mounds aligned along the two sides, and by two mounds piled up at its ends. The topography at the ground of the groove appears modulated as well. The height difference between the top of the mounds and the bottom of the pits in the groove is about 2.5 nm. The distance between two adjacent pits or mounds is about 40 nm, as well as the distance between the two rows of mounds.

Figure 1(b) compares a cross section of the topography along the ground of the groove with a measurement of the lateral force recorded while scratching the groove. The topographic image has been recorded with very low load on the tip after the scratching experiment was finished. The topographical information recorded simultaneously with the scratching cannot used for the comparison due to strong cross talk between lateral and normal force at the high load required for scratching. The lateral force is modulated with the same periodicity as the topography along the groove. Note that there is no stick-slip characteristic but a continuous sliding with modulated lateral force, i.e., with modulated velocity in the tip. Any increase of the lateral force is a direct indication for a slower tip movement with respect to the movement of the support. Comparing topography and lateral force in Fig. 1(b) we find that the lateral force starts to increase whenever the tip is at the edge of a pit between two mounds. The tip movement is decelerated at the slope of the mounds, and a lateral strain is built up. At a certain level the lateral force is high enough to pull the tip out of the pit and with some accelerated velocity the tip moves onto and over the mound. These results indicate that the formation of the regular ripples is related to an interplay between the developing topography and the lateral strain produced by the tip of the force microscope.

Figure 2 shows a series of grooves with a length of 800 nm produced by scratching the same surface 512 times with a velocity of 2.67 μm/s and with varying loads. There is a clear monotonous increase of the depth of the grooves and the height of the mounds piled up around the grooves with increasing load. However, the dependence of the development of undulated structures on the load is much less evident. Some grooves exhibit hardly any ripples at their edges, and some only close to their ends. There is no clear tendency in the effectiveness of ripple formation depending on the applied load. However, in numerous experiments we found that such ripples form for all loads and velocities after prolonged scratching. To summarize these observations, we find that wear always increases with load, while the ripple formation is a more irregular process which, however, after some time always starts.

In order to visualize the onset of ripple formation, Fig. 3 shows the lateral force recorded while scratching along one line as a function of time. For the first few scan lines, the friction force is low and no structure can be recognized. Wear starts then at a point in the center of the scan line, and spreads out first to the right and then to the left. The higher friction is caused by the enhanced interaction of the tip sliding in the groove compared to sliding on an atomically smooth terrace. The friction force is developing some faint structure, which is again lost in the course of further scanning. After about one third of the time represented in this figure, another instability similar to the one starting the whole process in the beginning is observed. Taking into account the depth of the final groove of about 0.6 nm [cf. Fig. 1(b)], it is plausible to assume that wear of a second atomic layer starts at that point. Now the ripple pattern in the lateral force evolves and is stable throughout the experiment. Unfortunately, we cannot quantify lateral force and topography at the same time. In order to avoid any disturbance of the lateral force measurement[9] by the distance feedback, the latter is extremely slow and maintains only a mean constant normal load.

It is worth mentioning that the lateral force is relatively strong and irregular each time after a new atomic layer starts to wear off. This is documented in the corresponding graph

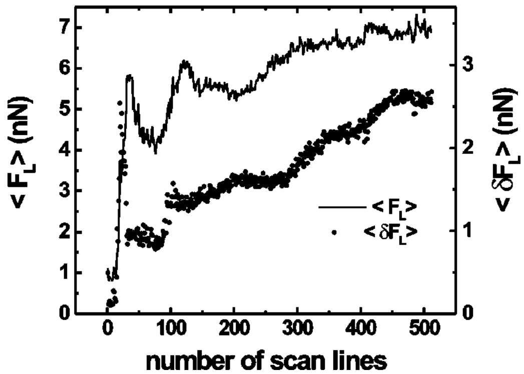

FIG. 4. The development of the mean lateral force $\langle F_L \rangle$ and of its standard deviation $\langle \delta F_L \rangle$ while producing the ripple pattern shown in Fig. 3.

in Fig. 4. The mean lateral force $\langle F_L \rangle$ shows steplike increases, with a strong peak at the position of the first step and a less pronounced peak at the position of the second. For longer scratching we have found that the mean lateral force slowly levels off.

The standard deviation of the lateral force $\langle \delta F_L \rangle$ exhibits steplike increases where a new atomic layer is eroded, reflecting the growing amplitude of the lateral force when the worn-off atoms are reorganized in form of the ripples. Judging from this graph, we might even identify the erosion of a third layer around the 280th and of a fourth layer around the 400th scan line. In repeated longer experiments we have not observed a saturation of the increase of the ripple amplitude. Again, the onset of erosion of the first two atomic layers is indicated by sharp peaks which reflect the irregularity of the atomic reorganization process.

The distances between the tip-induced ripples on KBr increase slightly with an increase of the applied normal load. However, they vary significantly between experiments using different tips. Lateral force curves of the stable ripple pattern obtained in two different series of experiments are compiled in Fig. 5. While the typical features described above are reproduced for all tips and samples, the ripple distances vary from 20 nm to 50 nm. This length scale matches the typical radii of force microscope tips. Therefore, we speculate that the distance between the ripples is correlated with the actual tip shape. The increase of the ripple distance with load suggests that the increasing contact area as well influences the ripple distance. Note that in Fig. 5 the curve with larger ripple distance shows higher values of the lateral force, indicating again a larger contact radius.

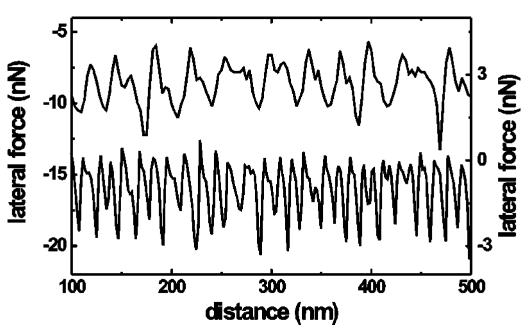

FIG. 5. Lateral force recorded during the 1250th scratch using two different tips and samples (upper curve is quantified by the left scale, lower curve by the right one). The normal load (3.6 nN) was the same for both curves.

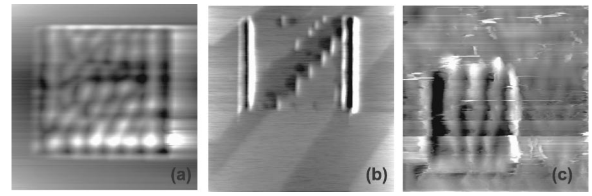

FIG. 6. Topography images of square areas scanned before with higher load. (a) Flat KBr(100) surface (sample A). The eroded square has a side length of 500 nm. (b) KBr surface with cleavage steps running from the lower left to the upper right corner of the frame (sample B). The eroded square has a side length of 500 nm. (c) Al(111) surface with an eroded area of 400 nm side length.

Two-dimensional patterns in the lateral force with similar periodicity are obtained if the tip is scanning a square, by slowly moving along the (010) direction while scanning forwards and backwards along the (100) direction. Figure 6(a) shows the result of scanning 34 times a square frame of 300×300 nm^2, composed of 256 horizontal lines. The vertical distance between two consecutive scan lines is much smaller than the typical width of the grooves observed in Fig. 2. A pattern of ripples is revealed, with a preferential alignment perpendicular to the fast scanning direction of the tip. The typical wavelength of the ripples is about 40 nm. On the surface cleaved in UHV (sample B) the initial stages of the ripple formation were influenced by the cleavage steps [Fig. 6(b)]. The steps are eroded and aligned perpendicular to the fast scanning direction, and serve as nucleation sites for the first ripples. After prolonged scanning, however, the pattern becomes independent of the original morphology and adopts a shape comparable to Fig. 6(a).

The phenomenon of ripple formation in friction force microscopy experiments on the nanometer scale occurs on a variety of surfaces. Besides polymer surfaces[4] metal surfaces may also develop regular topographic features. In Fig. 6(c) we present a similar result, which we obtained on an Al(111) surface. Again, a two-dimensional pattern of ripples evolved under the scanning tip. However, this surface is very sticky, and even at lowest normal load all topography images show significant disturbance by sticking effects. In contrast to KBr, the ripple pattern on Al(111) is unstable under prolonged scanning and turns into an irregular structure related to very strong wear processes.

III. DISCUSSION

In the following, we will discuss the tip-induced process of ripple formation along the results presented in the preceding section. First of all, this process starts only after the onset of abrasive wear. No signs of topographic modulation have been observed without an erosion of at least one monolayer of KBr, i.e., without transfer of material. Elastic instabilities or plastic deformation related to creation or movement of dislocations can be excluded, when the strain energy produced by the scratching tip is smaller than the energy required for the creation of dislocations. Based on our observations, we believe that this is the case in our experiments. Similar arguments have been brought forward in the discussion of nanoscale indentation of metals.[10,11]

Once an atomic layer is damaged at some specific spot, the whole layer is removed and its material reorganized in a process accompanied by strong, irregular friction. After some tens of scan lines (compare Fig. 4) a more ordered structure is formed. This process was described in detail in a previous study,[12] where it was shown that the material is continuously detached in small amounts of ion couples or small clusters. The debris at the end of the scratch, i.e., in front of the moving tip, perfectly recrystallizes in atomically flat terrace which exhibits exact atomic registry with the underlying substrate lattice. Some understanding of the process can be obtained from comparing the corrugated topography and lateral force profiles in Fig. 1. The ripples probably emerge when material transported in front of the tip increases the friction, slowing down the tip movement until the lateral force is strong enough to make the tip jump over the mound of material. During following scan lines, existing mounds will collect further material and grow. In this scenario the minimal distance between two ripples would be given by the size of the tip, in agreement with the experimental findings.

In a recent paper Friedrich *et al.* investigated the action of a localized moving disturbance which deposits or removes material from a surface.[13] They assumed that the surface profile evolves according to a generalized Kuramoto-Sivashinsky equation, which takes into account the inhomogeneity of the disturbance. The competition between a negative surface tension ν, which tends to increase the area of the surface, and a positive coefficient κ, which accounts for the surface diffusion, leads to a periodic structuring of the surface with wavelength $\lambda \sim \sqrt{\kappa/\nu}$. Similar mechanisms have been previously recognized as responsible for the ripple formation observed under ion sputtering,[14] although in such a case the perturbation is not localized, but acts on the whole surface at the same time. In our case the negative surface tension can be identified with the stronger erosion in the pits connected with the higher strain exerted by the trapped tip. On the other hand, the perfect recrystallization of the piled-up mounds indicates that diffusion also plays a smoothing role for the ripple formation on KBr.

It is interesting to note that thin films of KBr produced by molecular evaporation grow in form of pyramidal mounds, where the topmost terraces have typical sizes in the range of 20–50 nm,[15] i.e., in exactly the same size range we find for the ripple distances. In the present results, the relation between height and width of the mounds is low compared to typical growth pyramids and, therefore, diffusion does not limit the further increase of the surface corrugation. This is reflected in the lack of saturation of the lateral force variation in Fig. 3. Based on the experimental results, we cannot rule out the dominant parameter for the distance of the ripples. The weak dependence of the distance on the normal load may suggest that both a characteristic diffusion length and the shape of the tip might be important for the size of the developing structures.

IV. CONCLUSION

In conclusion, we have given a detailed experimental report on the formation of regular topographic structures on a KBr surface upon scanning a force microscopy tip in contact with the sample. Following abrasive wear of single atomic layers, the debris is moved and reorganized in an interplay between friction-induced strain and erosion, transport of material by the action of the tip, and possibly diffusion. The self-amplification of the evolving structures has an important impact on the tribological behavior, where stick-slip phenomena on different length scales are a general observation.

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