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Nanostructure formation due to impact of highly charged ions on mica

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A B S T R A C T

Muscovite mica was irradiated with slow highly charged Ar^{q+} (charge state $q = 12, 16$) and Xe^{q+} ($q = 23, 27$) ions in a kinetic energy range of 150–216 keV and subsequently observed by contact mode atomic force microscopy. Surprisingly, on samples irradiated with Xe ions nano-sized hillock-like structures were found well below the charge state threshold reported in earlier experimental investigations. However, the structures found are not the result of a true topographic surface modification induced by the ion bombardment, because the absence of these nanostructures in tapping mode images and the dependence of the detected structures on scan conditions points towards a surface modification which manifests itself only in frictional forces and therefore in height measurement artifacts. Furthermore the generated defects are not stable but can be erased by continuous scanning.

1. Introduction

Slow (eV to keV) highly charged ions (HCI) have been suggested as a novel tool for gentle nanostructuring [1–3] of surfaces and meanwhile a variety of materials have been investigated by scanning probe methods [4,5]. Systematic studies in which charge state and kinetic energy of the HCI have been varied, however, have so far only been carried out for CaF_2, KBr, HOPG and Mica (see [4–6] and references therein).

In the case of CaF_2 [7–9], for example, it has been shown by decelerating the HCI projectiles to low kinetic energies ($150\,q \times$ eV) that the potential energy of a single ion alone can be sufficient to create a topographic, non-erasable nano-sized protrusion (hillock) on the surface. This holds true above a certain threshold of potential energy (around 12 keV), above which height and diameter of the hillocks increase with increasing potential energy. By simulations on the basis of an extended classical over-the-barrier model the observed threshold could be successfully linked to a solid-liquid phase transition (nano-melting) [4,7,10].

On KBr(001) surfaces bombarded with slow highly charged Xe ions, on the other hand, pit structures with lateral sizes of 10–25 nm and monoatomic depth are created above certain thresholds for both potential and kinetic energy [11]. The mean pit volume shows a linear dependence on the potential energy of the ions. Potential sputtering [12] by a defect-mediated desorption mechanisms was invoked to explain these results [11].

For HOPG nano-sized hillock-like structures were found by scanning tunneling microscopy (STM) for all projectile charge states and kinetic energies [13–19]. While the structure size increases with the potential energy of the ions [4,20], no pronounced dependence on the kinetic energy was found [18]. However in atomic force microscopy (AFM) measurements, the dependence of the detected structures on scan conditions points towards a surface modification which manifests itself only in frictional forces and therefore in height measurement artifacts [20]. Furthermore the generated defects are not stable but can be erased by continuous scanning in contact mode [20].

In this paper we present recent results for HCI-induced nanostructures on muscovite mica, a phyllosilicate with the sum formula $KAl_2(Si_3Al)O_{10}(OH)_2$, which consists of layers with a net negative charge bonded through interlayer cations. This material is known to be a very good insulator and simple sample preparation by cleaving with adhesive tape or razor blades gives atomically flat surfaces with occasional occurrence of atomic steps. Mica is commonly used as a substrate for AFM investigations, or, as a calibration material and is known to be stable under contact mode atomic force microscopy. Due to these beneficial properties, mica has been considered a favorable material to explore ion-induced modifications.

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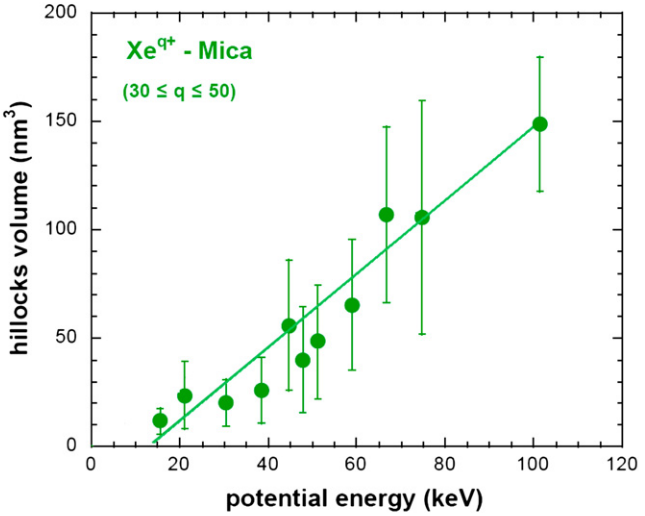

Fig. 1. (colour online): Mean hillock volume as a function of potential energy on mica after irradiation with Xe^{q+} ($30 < q < 50$) ions. Data taken from [21] and [28].

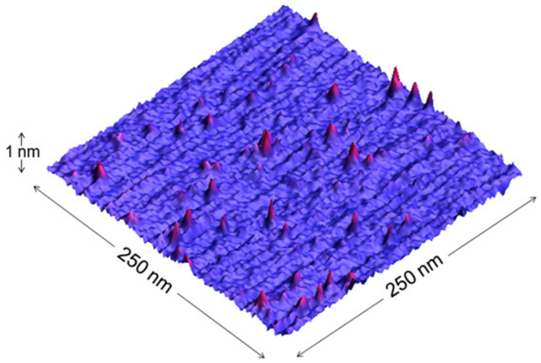

Fig. 2. (colour online): Topographic contact mode AFM image (250 nm × 250 nm) of a mica surface irradiated by 150 keV Xe^{23+} ions showing hillock-like nanostructures protruding from the surface as a result of individual ion impacts.

In section 2 we first summarize the results of previous studies on slow highly charged ion irradiation of mica. After a description of the experimental methods used in section 3, we present our latest results obtained by atomic force microscopy (AFM) in section 4.

2. Previous work on mica

First studies on the influence of slow HCI impact on mica surfaces were performed by the group of D. Schneider at Lawrence Livermore National Laboratory [21–23]. The cleaned and cleaved mica surfaces were bombarded with HCI (e.g. Kr^{35+}, Xe^{35+}, Xe^{44+}, Th^{74+} and U^{70+} ions) extracted from an EBIT (electron beam ion trap) source at typical energies of 2.2 keV/amu and total ion fluences of $\sim 10^9$ ions/cm^2. AFM investigation performed under ambient conditions in contact mode revealed hillock-like protrusions for Kr^{35+}, Xe^{44+}, Th^{74+} and U^{70+} ions, but no structures were observed for Xe^{35+} and singly charged Xe^{+} projectiles. By comparing the number of defects found to the total ion fluence, each hillock was found to be the consequence of a single ion impact. The structures were called “blisters” because sometimes, after continuous scanning of a “blister” by contact AFM, craters were observed. As there was no change in the appearance of the structures upon reversal of the scan direction, their origin was ascribed to a true topographic surface modification and not related to changes of the friction coefficient as in the present and other studies (see below).

In an extension of their work, D. Schneider and co-workers [24,25] correlated the volume of the blisters with the charge of the incident ion, with a threshold for blister formation at a charge state of $q = 35$. The diameter of the blisters was found to increase from 10 to 40 nm with incident charge from 35+ to 70+. Two theoretical models were proposed to explain the mechanism of defect creation. One is based on the Coulomb explosion model [26], the other associates defect production with the enclosure of neutral entities, indicated by the observation that similar defects are caused by heating of the mica substrate. Heating could lead to expansion of the trapped volumes or to disturbance of the charge balance by displacement of the interlayer cations.

Parks et al. [27,28] have repeated these measurements. At a constant kinetic energy (100 keV) of the incident Xe ions they observed a similar blister volume dependence on the incident ion charge, with a threshold at $q = 30$. A summary of the obtained mean hillock volumes for investigated Xe charge states from [21] and [28] is depicted in Fig. 1. In addition Parks et al. [27,28] confirmed that, under the special conditions they used, the blister size is independent of the kinetic energy (20–800 keV) of the highly charged projectile. The researchers assumed that hillock creation is closely related to the layered structure of mica. Adjacent sheets may be easily forced apart by coulomb repulsion due to the rapid removal of electrons in the near-surface region. Locally this may be seen as a protrusion. This interpretation is also consistent with the observation that the hillocks were not stable over time but often disappeared after continuous scanning over the same area. Similar to the behaviour found in the present study (see section 4), the researchers observed that, depending primarily on loading force and scan angle, the defects may appear either as hillocks or as pits in the AFM topographic image when the scan direction is reversed. However, as pits were also visible in tapping mode AFM imaging, where lateral forces are minimized, the authors propose that the structures formed are due to a true topographic modification and that the contrast reversal seen in contact mode is a consequence of both topographical and frictional contributions.

A study on the track formation induced by swift heavy ions on mica [29] found the same scan-angle dependent appearance of ion-induced structures. However, here a comparison of images recorded at different scan angles led to the conclusion that direction-dependent friction forces gave rise to artificial topographical features and that the observed contrast originated solely from friction.

3. Experimental methods

Irradiations of the mica samples were carried out at the ARIBE [30] facility of GANIL (Grand Accélérateur National d'Ions Lourds) in Caen, France. Xe^{q+} ($q = 23$, 27) and Ar^{q+} ($q = 12$, 16) ions were extracted from a 14.5 GHz ECRIS and accelerated onto the samples under normal incidence to final impact energies of 150–216 keV. For every desired charge state two freshly cleaved samples were fixed on a target holder and mounted in the target chamber at pressures of $\sim 10^{-7}$ mbar. Time averaged beam fluxes of 10^9–10^{10} ions/s and irradiation times of 20 sec up to 10 min resulted in total ion fluences from several 10^9 ions/cm^2 up to 10^{11} ions/cm^2. For each charge state, two samples were irradiated with rather different fluences (typically a factor of 5 difference) to be able to unambiguously ascribe observed nanostructures to (single) ion bombardment. Samples were investigated immediately after irradiation with a Nanoscope III (Digital Instruments) AFM in constant force contact mode operated under ambient conditions with triangular Si_2N_3 cantilevers