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AFM studies of a new type of radiation defect on mica surfaces caused by highly charged ion impact

C. Ruehlicke *, M.A. Briere, D. Schneider

Lawrence Livermore National Laboratory, Livermore, California 94550, USA

Abstract

Radiation induced defects on mica caused by the impact of slow very highly charged ions (SVHCI) have been investigated with an atomic force microscope (AFM). Freshly cleaved surfaces of different types of muscovite were irradiated with SVHCI extracted from the LLNL electron beam ion trap (EBIT) at velocities of about 2 keV/amu. Atomic force microscopy of the surface reveals the formation of blisterlike defects associated with single ion impact. The determined defect volume which appears to increase linearly with the incident charge state and exhibits a threshold incident charge state has been determined using the AFM. These results indicate that target atoms are subjected to mutual electrostatic repulsion due to ionization through potential electron emission upon approach of the ion. If the repulsion leads to permanent atomic displacement, surface defects are formed.

1. Introduction

Radiation defects in solids have long been observed with nuclear detectors in the form of tracks in the material bulk, caused by impact of high energy (MeV) charged particles. Presently, much effort is devoted towards the understanding of the interaction of SVHCIs with solid surfaces. With the recent development of electron beam ion traps (EBIT), which allow the extraction of highest charged ions at kinetic energies in the keV range, the formation of surface defects has been made possible [1–3]. Investigation of the underlying mechanisms of surface defect formation is of interest for understanding the basic physics of ion solid interactions as well as for possible nanometer scale lithographic applications due to ion induced surface modifications.

The LLNL EBIT 2 provides ions with charge states up to $q \approx 80+$ which carry up to ≥ 200 keV potential energy. Atomic force microscopy makes it possible to examine the defect topography on a nm scale qualitatively and, to a lesser extent, quantitatively. The latter, however, is still limited due to uncertainties in the AFM tip–surface interaction and tip shape effects.

2. Experimental

The ions used for irradiation of the targets were extracted using the LLNL EBIT 2 [4,5]. It generates highly charged ions by successive electron impact ionization from an electron beam: atoms from a gas source or lowly charged ions from a metal vapour vacuum arc source (MeVVA) are injected into the trap volume, where they are radially confined by the space charge of the electron beam while a drift tube assembly, consisting of three differently biased segments, provides axial confinement. To extract the ions, the potential of the middle drift tube is raised so that the ions can overcome the potential well. The charge state distribution of the extracted ions is determined by the confinement time, i.e. the time they are subjected to the electron beam impact. The target material is 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 structure results in an excellent cleavage behaviour providing very flat surfaces.

Freshly cleaved samples of muscovite mica were irradiated with different ions; Xe^{1+}, Xe^{35+}, Kr^{35+}, Xe^{44+}, Xe^{50+}, U^{70+} and Th^{74+}, extracted at a velocity of 2.2 keV/amu and fluences of 10^9–10^{10} ions/cm^2. The vacuum along the beam path and in the target area was about 5×10^{-9} Torr. The ion flux was measured with a detection system consisting of a microchannel plate array or photomultiplier, with an ion detection efficiency between

* Corresponding author. Also Universität Bielefeld, 33615 Bielefeld, Germany.

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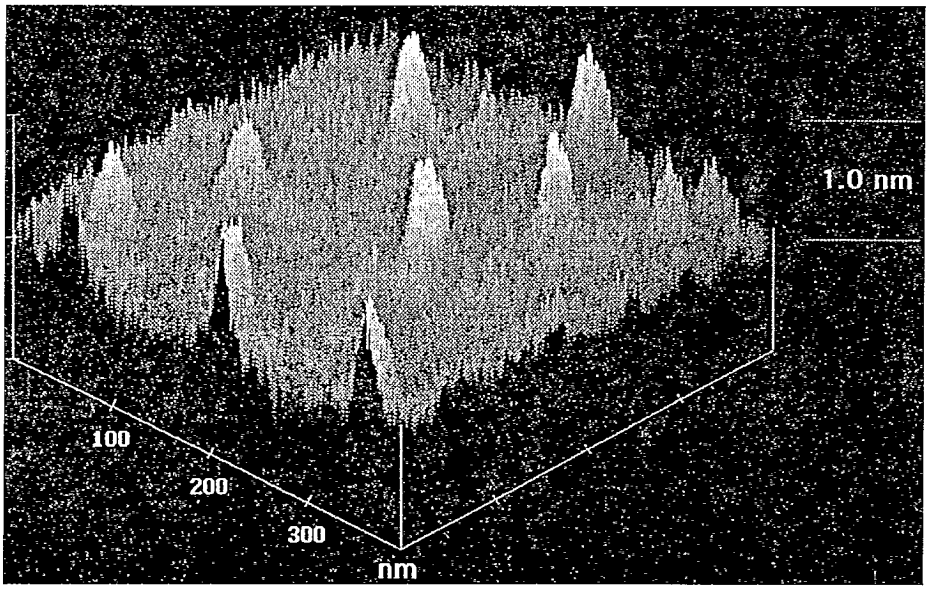

Fig. 1. AFM image of mica irradiated with slow U^{70+} ions. The perspective view shows blisterlike defects with a full diameter of 30 to 50 nm and a width of 0.5 to 1 nm.

30 and 60% depending on the details of the actual setup. The fluence was typically achieved in a 1–10 h irradiation, depending on the ion species.

A series of irradiations was performed at different velocities: 1.3, 2.2 and 4.5 keV/amu. After irradiation the targets were stored in air until being imaged with the AFM.

3. AFM of the irradiated surfaces

The irradiated samples were imaged with two different commercial AFMs: Nanoscope III (Digital Instruments) and Autoprobe LS (Park Scientific Instruments). The AFM images topographical structures of a surface by scanning it with a very sharp tip attached to a cantilever. The bending of the cantilever is a measure of the height change on the surface and is monitored via a laser beam, reflected off the cantilever onto a photosensitive detector. An electronic feedback loop is used to adjust the height of the sample at every point, thus keeping the cantilever deflection constant. With the Nanoscope III a pyramidal Si_3N_4 tip of aspect ratio 1:1 was used which was attached to a cantilever with a nominal rigidity of 1 nN/nm. All of the samples were measured with the Nanoscope III shortly after the irradiation. Results for height and width of the defects were obtained by assuming a linear background and measuring the full width w and height h of the defect with respect to that background. Assuming a spherical shape of the defects, the volume was then obtained by: $V = w^2h/2$. The noise on both instruments is still on the order of a few Å, so that very small defects, with a volume less than 10 nm^3, according to a rough estimate, cannot be detected.

4. Results and discussion

Observation of the sample surface showed that blisterlike defects had been generated during irradiation (Fig. 1). Their shape is independent of the scanning direction (Fig. 2) indicating real topographic structures, rather than features due to alteration of the friction coefficient [6]. The density of the defects, which is comparable to the ion fluence, indicates that they are caused by single ion impact. They range from 0.1 to 3 nm in height, although most vary between 0.5 and 2 nm. The width appears to be

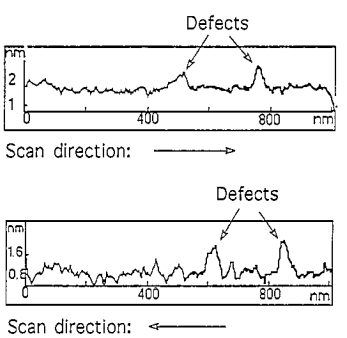

Fig. 2. Line section of two AFM scans of mica irradiated with Xe^{50+} ions. The scans, depicting the same two defects, were taken with opposite scan directions, showing that the defect appears as a blister independent of the scan direction.

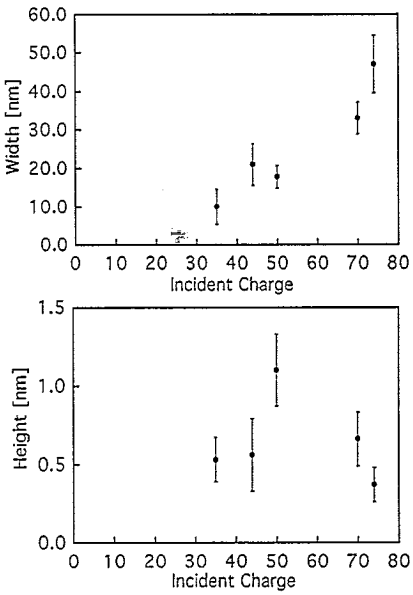

Fig. 3. Mean heights and widths of defects as a function of incident charge state determined with Nanoprobe III. The error bars represent the statistical width of each distribution.

on the order of 10 to 60 nm. Figs. 3 and 4 show the height, width and volume of the defects caused by different incident ions as measured with the Nanoscope III. The dimensions obtained with the Autoprobe LS vary from the Nanoscope III results, which might be partly ascribed to the use of more brittle Si tips with the former instrument. The height measurement is expected to be sensitive to the force exerted by the AFM cantilever, it is therefore possible that the defects were squeezed down. The estimated volume, however, increases linearly with charge state of the incident ion (see Fig. 4).

No defects were seen on mica irradiated with Xe^{1+} or Xe^{35+}. The defect size was not found to depend on the velocity of the projectile, which had been varied over the range of 1.3–4.5 keV/amu. Mica samples of different composition seem to yield different size defects, a systematic study of this effect is underway. Fig. 5 shows the

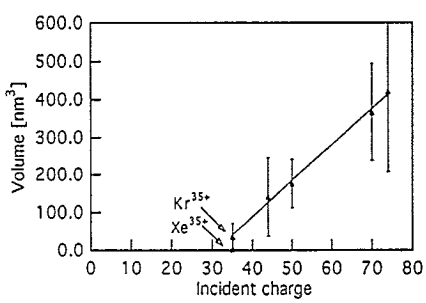

Fig. 4. Mean volumes of defects as a function of incident charge state determined with Nanoprobe III. The error bars represent the statistical width of each distribution.

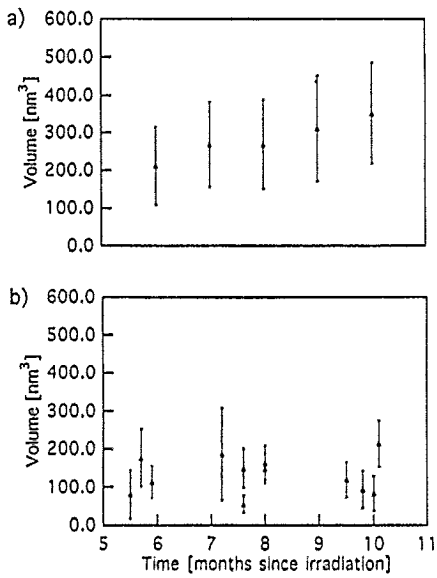

Fig. 5.(a) Mean volume distribution of defects caused by Xe^{44+} ions on various mica samples during different independent experiments. The error bars represent the statistical width of each individual distribution. (b) Mean volume measurement results from one sample irradiated with U^{70+} ions measured several times over a period of time. The error bars represent the statistical width of each individual distribution.

stability of the defects over time and the reproducibility of the irradiation. The volume measurement has been reproduced within 5%.

The mechanism by which the defect formation is governed is not yet understood. One model follows the ion explosion spike model by Fleischer et al. [7] which has been introduced to explain the formation of nuclear tracks. The electrical field of the ion approaching the surface draws electrons out of the solid thereby charging it positively. The ions in this charged regions repel each other and, if the repulsive force exceeds the mechanical strength of the material, permanent atomic displacement occurs. A minimum threshold charge state is expected, which should be around $q \sim 30$, according to experimental observation. Considering the structure of mica, one expects the top layers to mutually repel each other and form a hollow blister, which would appear as a hillock during AFM measurement. The amount of charge of the incident ion then influences the strength of the electric field drawing electrons from the solid which in turn determines the size of the affected surface region. Therefore one expects the defect volume to increase with increasing charge state. In the case of conductors the increase of the electron emission with incident charge has been measured [8]. Defect production may also be associated with enclosure of neutral entities, indicated by the observation that similar defects are caused by heating 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. Although first investigations have shown formation of similar defects through SVHCI irradiation and heating it is not clear whether the same mechanism is responsible in both cases or whether they are of a different nature.

5. Conclusion

Defect formation due to irradiation of muscovite mica with SVHCI has been described. Although AFM investigation of the defects has revealed an effect of the incident charge state on the defect size which increases with increasing projectile charge there are still large uncertainties in the absolute dimensions of the defects involved and the underlying mechanisms are not yet understood. It has been shown that defects are not uniquely caused by irradiation but are also generated by sample heating. The studies are to be extended to AFM investigation on a sub nm scale as well as systematic irradiation of different kinds of mica.

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