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ЯРОСЛАВСКИЙ ГОСУДАРСТВЕННЫЙ УНИВЕРСИТЕТ
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Том 1

Nano-hillock formation on CaF_2 due to cluster-ion irradiation

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Slow highly charged ions (HCIs) and swift heavy ions (SHIs) are known to induce nano-structuring on semi-conductors and insulators due to electronic excitations in the target's electronic system. The bandgap of such materials results in an electronic confinement, preventing the energy depositioned in the target from dissipating. Subsequent electron-phonon-coupling starts a rapid thermal expansion and further quenching of the irradiated area, forming protruding nano-hillocks on the surface [1-5]. This process is shown in figure 1 on the left side.

We present here a direct way to generate nano-hillocks by irradiating a CaF_2 (111) surface with slow heavy cluster ions, triggering the hillock formation by deposition of the ions' kinetic energy into a small volume directly into the targets' lattice, which can be seen schematically in figure 1 on the right side. CaF_2, an earth-alkalide halide, was repeatedly

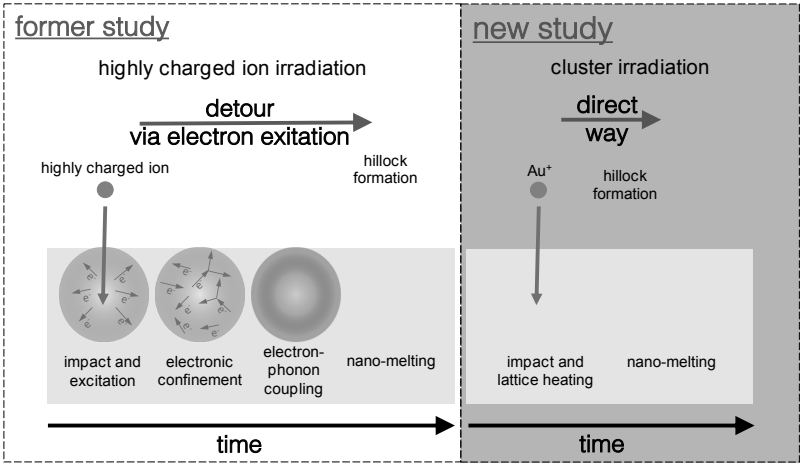

Figure 1 Schematic of processes inducing nano-hillock formation, describing the indirect process via electron-phonon coupling on the left side and the direct process via direct knock-on on the right side.

investigated regarding nano-structuring with SHIs [4,5] and HCIs [1-3] and therefore provides a data set to draw direct comparison with our new study.

The samples were investigated with AFM in tapping mode under ambient conditions, revealing atomically flat surfaces with mono-atomic steps that are a result from the production process. Some areas were also found to have clustered islands that appear to be remaining contaminations on the surface. The irradiations were performed at the Ion Beam Center of the Helmholtz-Zentrum Dresden-Rossendorf (HZDR) utilizing a liquid metal alloy ion source [6] with $Au_{82}Si_{18}$ as source material. Gold clusters Au_a with sizes a = 1-10 were extracted and accelerated to 30 keV. To differentiate a possible nano-hillock formation from contamination, different fluencies were used for irradiation, ranging from $1*10^{10}$ up to $1.3*10^{12}$ ions/cm^2. For better reproducibility of the analysis, the irradiations were performed on two different samples on different areas, where the irradiated surface is comparable to the pristine one on each sample. AFM tapping mode measurement under ambient conditions was performed after irradiation at the TU Wien.

Hillock-like nanostructures, using a Cypher AFM with Olympus Micro Cantilever OMLC-AC240TS-R3 tips (tip constant 2N/m) and a nominal tip radius of 7-12 nm, are comparable

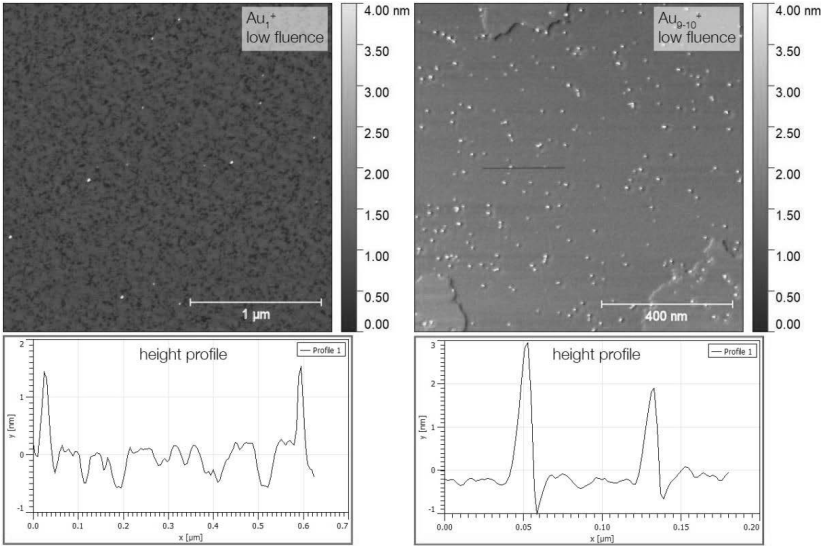

Figure 2 AFM tapping mode image of irradiated CaF2, including height profiles of nano-hillocks for irradiation with Au_1^+ and $Au_{9\text{-}10}^+$ ion cluster.

to those found in [1,2]. Figure 2 shows topographic AFM images of a CaF_2 (111) surface irradiated with Au_1^+ (a) and Au_{9-10}^+ (b), both with fluencies of about $1*10^{10}$ ions/cm^2. With increasing fluence the number of nano-hillocks increases, pinpointing the origin of those structures to the cluster-ion irradiation. To get sufficient statistics, about 100-150 hillocks per measurement were analysed regarding their height and volume. The areal density of nano-hillocks generated was found to be in the range of 20% of the applied fluence, which may result from systematic uncertainties in fluence determination in a FIB with very low fluencies compared to typical FIB experiments.

As it can be seen in figure 2, the height of nano-hillocks was found to be in the range of 1-2 nm. For Au_1^+ the mean height is 1 nm, while the mean height for Au_{9-10}^+ is about 2 nm, indicating an increase in hillock height with increasing cluster size. Comparing the results for Au_1^+, Au_2^+, Au_3^+, Au_{5-7}^+ and Au_{9-10}^+ an increase in cluster-height is observed. Assuming that the kinetic energy is distributed evenly to the atoms forming the cluster-ion after break-up, each atom has a specific stopping power that is related to its initial kinetic energy. Thus, larger clusters obtain lower stopping per atom but higher overall stopping per cluster.

To draw a comparison with existing results [1,2], first the creation of nano-hillocks induced by HCIs is discussed. An impinging HCI deposits a certain amount of energy into the target, which is a sum of electronic stopping, nuclear stopping and potential energy. While the first two parts are only dependent of the initial kinetic energy, the latter one depends on the charge state of the HCI. The nuclear stopping resembles the direct transfer of energy to the targets lattice system, which is a direct excitation of phonons. Contrary to this, the electronic stopping and the potential energy induce an excitation of the electronic system of the target. For HCIs, the electronic stopping was found to be negligible [1], therefore the potential energy is responsible for the formation of nano-hillocks on a CaF_2 (111) surface. When the HCI approaches the targets surface, an electron transfer from the target into the highly excited states of the ion starts, inducing a strong electronic excitation at the region of impact. Via electron-phonon-coupling, subsequent lattice heating, a rapid thermal expansion and further quenching of the target's material is induced, which is explained within the thermal spike model [7].
We propose, that for the irradiation with cluster-ions, the energy transfer solely takes place by direct knock-on processes and hence leading to an excitation of phonons in the target or direct atom displacements. The subsequent process of formation of nano-hillocks is assumed to be comparable to that found for nano-hillocks formed by irradiation with HCIs, namely the rapid

thermal expansion and quenching of the target material around the area of impact. The process of nano-hillock formation induced by cluster-ions is surface sensitive, i.e., it occurs only in the topmost layers of the target as the atoms in the cluster are effectively stopped in a few nanometres. It can be seen in figure 2 that the more atoms a cluster contains, the higher the nano-hillock gets. We assume that this is due to cluster-ions containing less atoms have a higher penetration depth into the target, triggering the phonon excitations at a deeper level and hence the thermal expansion takes place farther away of the targets surface. Figure 3 shows that the less kinetic energy per atom a cluster-ion obtained, the lower the interaction length/depth and the higher is the surface sensitivity becomes.

We see that it is not only possible to create nano-hillocks using cluster-ions, but that the height of the hillocks can be tuned by varying the size of the impinging cluster. Since we don't rely on a high bandgap target material for nano-structure formation, hillocks may be produced even on metallic surfaces, which are inert to SHIs and HCIs.

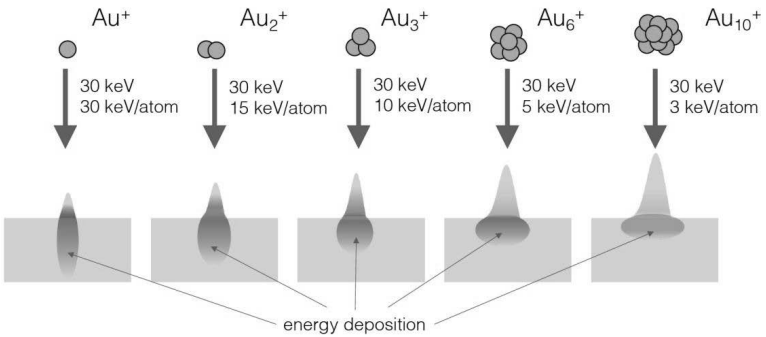

Figure 3 Schematic of the formation of nano-hillocks depending on the cluster-size and the kinetic energy per atom.

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