Surface defects in Al_2O_3, $MgAl_2O_4$ and MgO irradiated with high energy heavy ions

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Motivations

- Mechanisms of the surface defects induced by single swift heavy ion impact are not understood quantitatively
- Microstructural and surface effects of dense ionization in radiation-resistant ceramics and oxide crystals are of considerable practical value. To date, only a few data concerning the microstructural response of nonfertile ceramics to ion irradiation of fission energy are available. An external bombardment with energetic ions offers a unique opportunity to simulate fission fragment-induced damages

Aim of this work: the study of surface topography changes generated by (0.5 ÷ 5) MeV/amu Kr, Xe and Bi ions in Al_2O_3, $MgAl_2O_4$ and MgO crystals. Single ion induced effects are compared for set of ion fluences, ion incidence angles and irradiation temperatures.

Irradiation facility: ion beam line for applied research on the U-400 FLNR JINR cyclotron

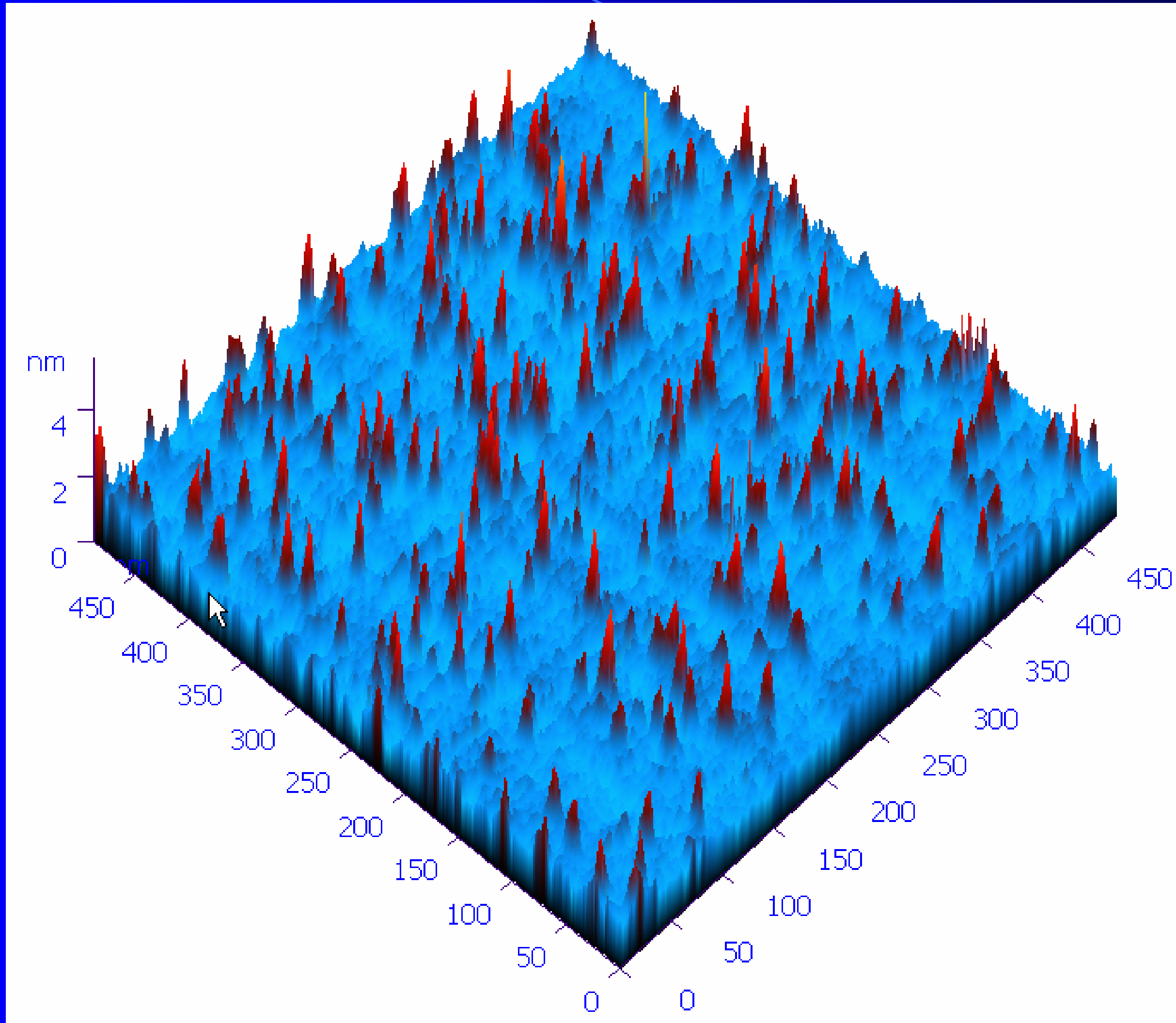
Ion beam homogeneity on irradiating specimen surface : 5%

ion flux density: < 2×10^{8} cm^{-2}s^{-1}.

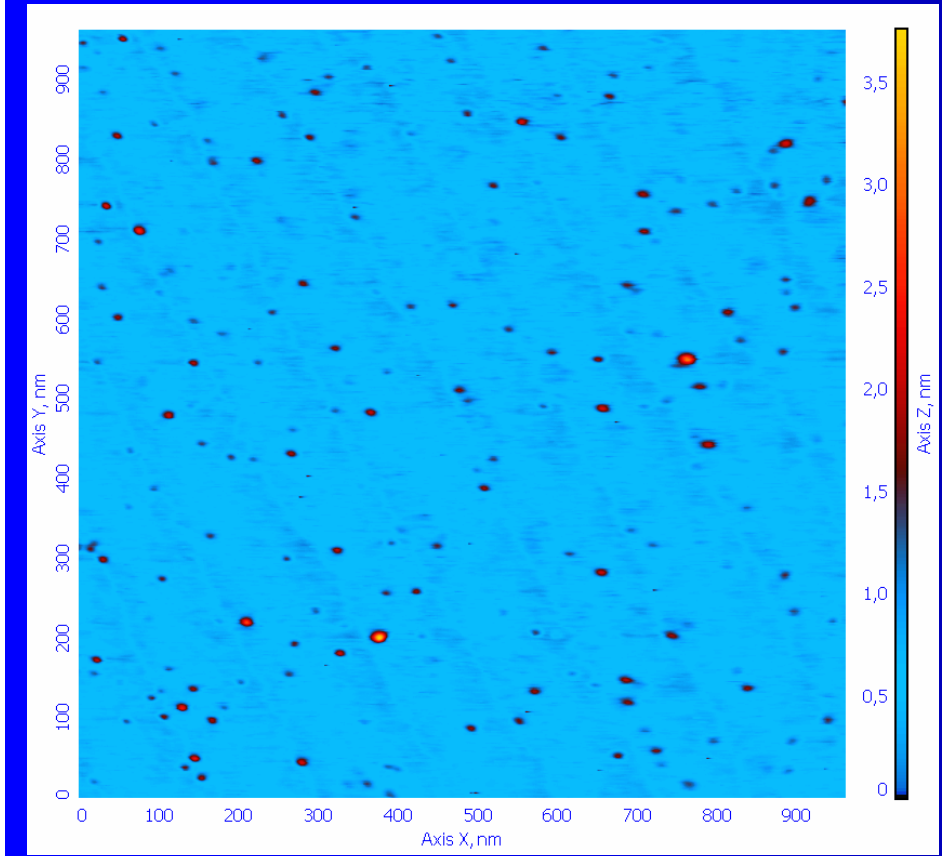
Characteristics of Bi, Xe and Kr ion irradiations

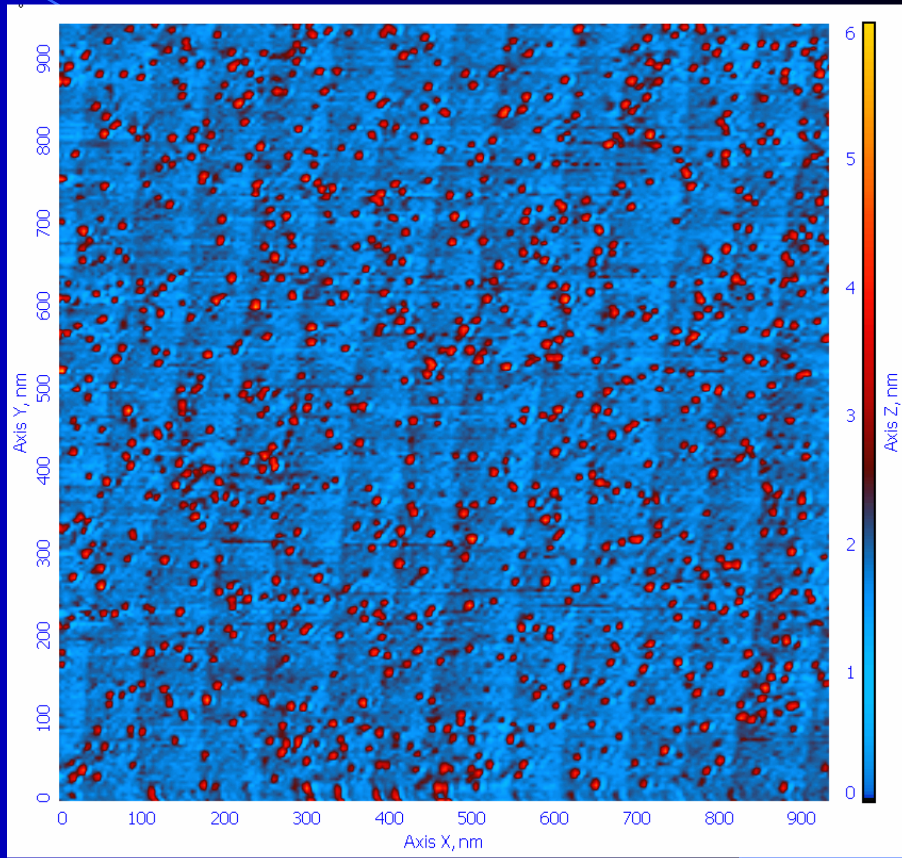
Ion type	Bi^{+51}	Bi^{+51}	Bi^{+32}	Bi^{+25}	Bi^{+17}	Kr^{+27}	Xe^{+38}
Ion energy, MeV	710	710	495	292	128	245	580
Target	Al_2O_3	$MgAl_2O_4$ MgO, SiC	Al_2O_3	Al_2O_3	Al_2O_3	$MgAl_2O_4$ Al_2O_3, MgO	$MgAl_2O_4$ Al_2O_3
S_e, keV/nm	41	34 36.7, 38.1	39	36	25.4	15.5, 16.8, 15.8	25 26.6
S_n, keV/nm	0.08	0.1	0.12	0.35	0.7	0.031	0.04
Incidence angle	0°, 30°, 45°, 60°, 75°	0°	0°	0°	0°	0°	0°
$T_{irr.}$, K	300, 78	300	300	300	300	300	300
Ion fluence, cm^{-2}	2×10^{10}, 10^{11} 5×10^{11}, 10^{12} 7×10^{12}	2×10^{10}	2×10^{10}	2×10^{10}	2×10^{10}	2×10^{10}	2×10^{10}

3D AFM image of of $MgAl_2O_4$ surface irradiated with 580 MeV Xe ions.
Ion fluence $1x10^{11}$ cm^{-2}



2D AFM image of spinel surface irradiated with 580 MeV Xe ions

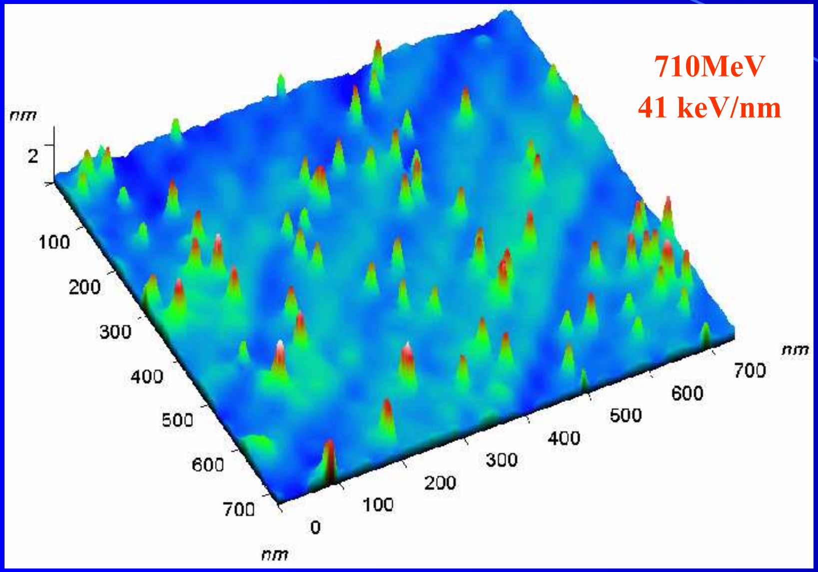


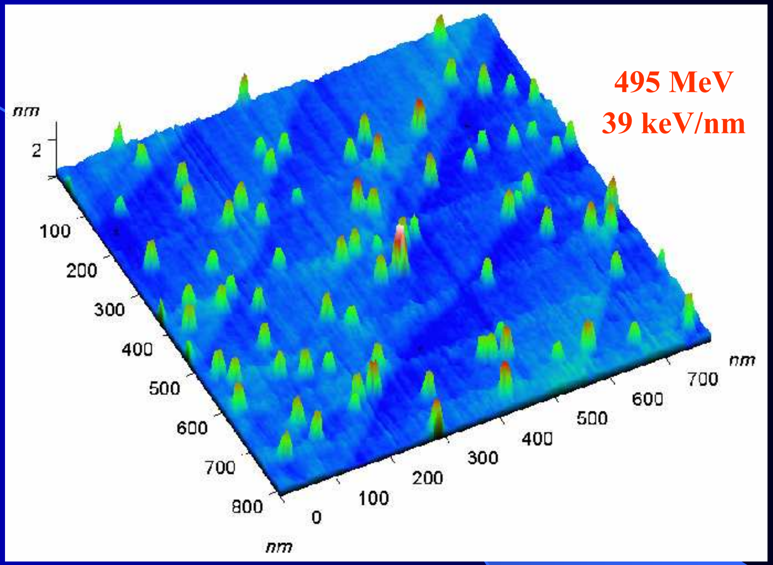


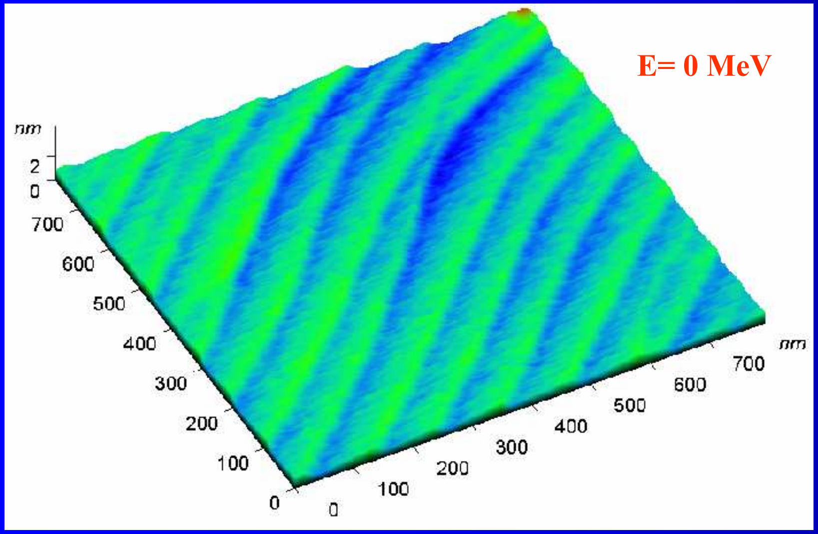
$\Phi t=2x10^{10}$ cm^{-2}
Number of hillocks: $1.87x10^{10}$ cm^{-2}

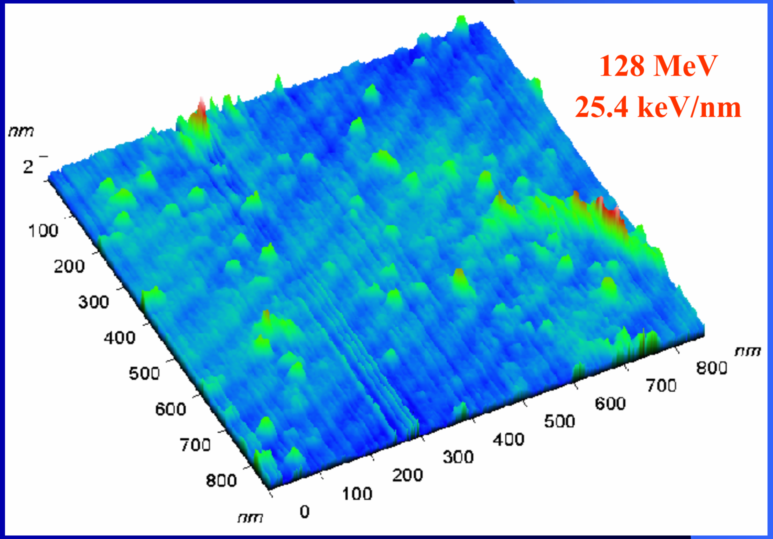
$\Phi t=1x10^{11}$ cm^{-2}
Number of hillocks: $0.91x10^{11}$ cm^{-2}

3D AFM images of of α-A_2O_3 surfaces irradiated with Bi ions at different incident electronic energy deposition

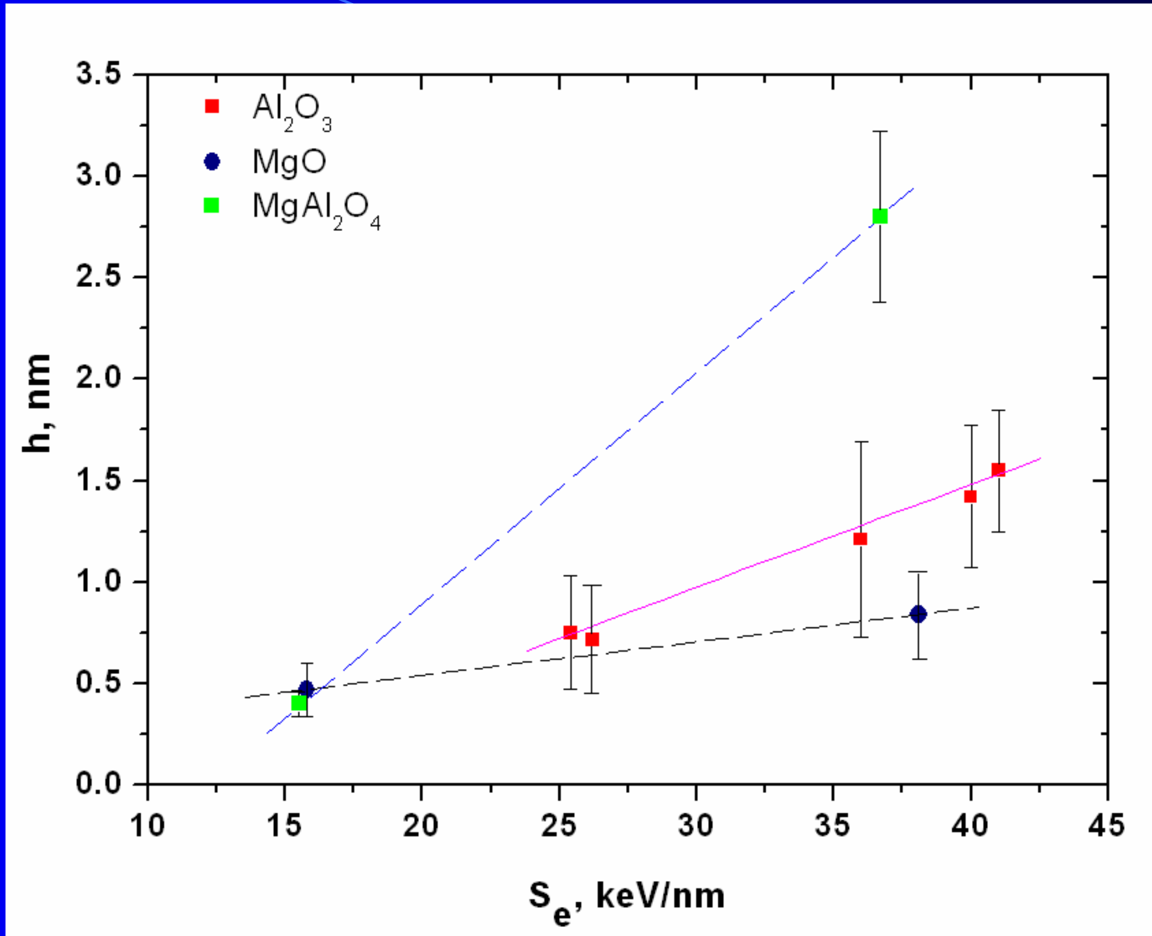








Mean hillock height versus incident electronic energy deposition

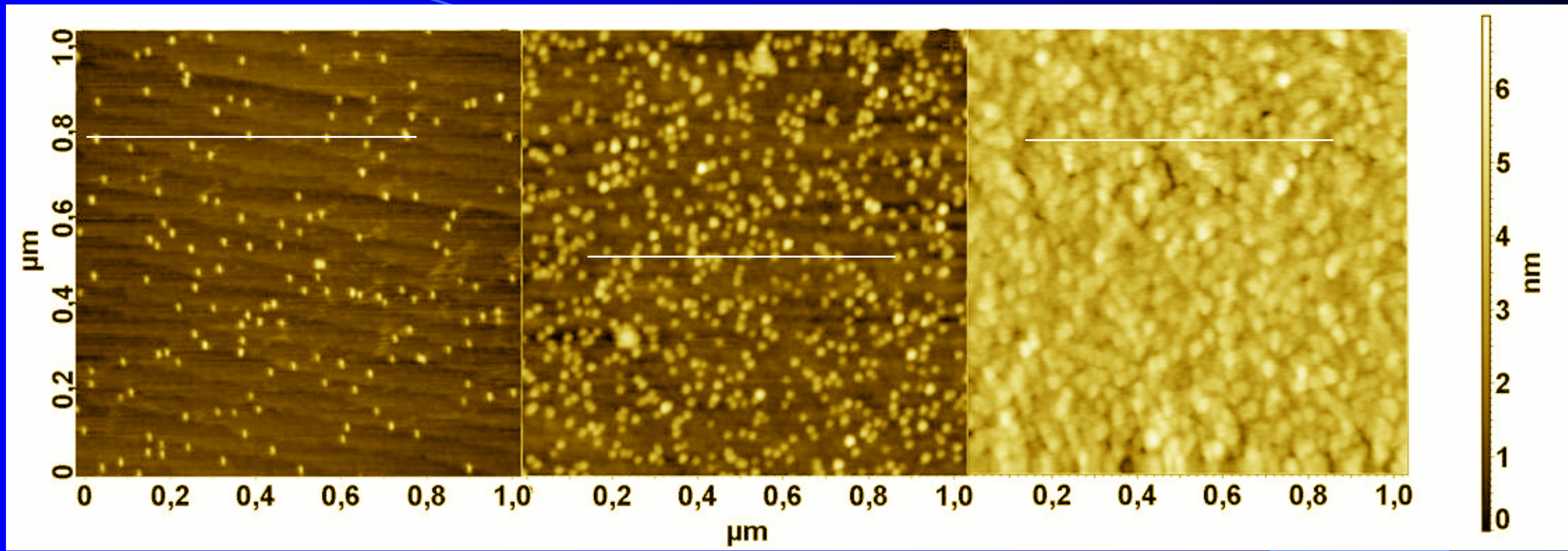


Threshold electronic stopping power value needed for the hillocks production:

MgO ≈ 15. 8 keV/nm, $MgAl_2O_4$ ≈ 15. 5 keV/nm, Al_2O_3 ≈ 25 keV/nm

SiC S_e > 34 keV/nm

A_2O_3 surface relief evolution with 710 MeV Bi ion fluence

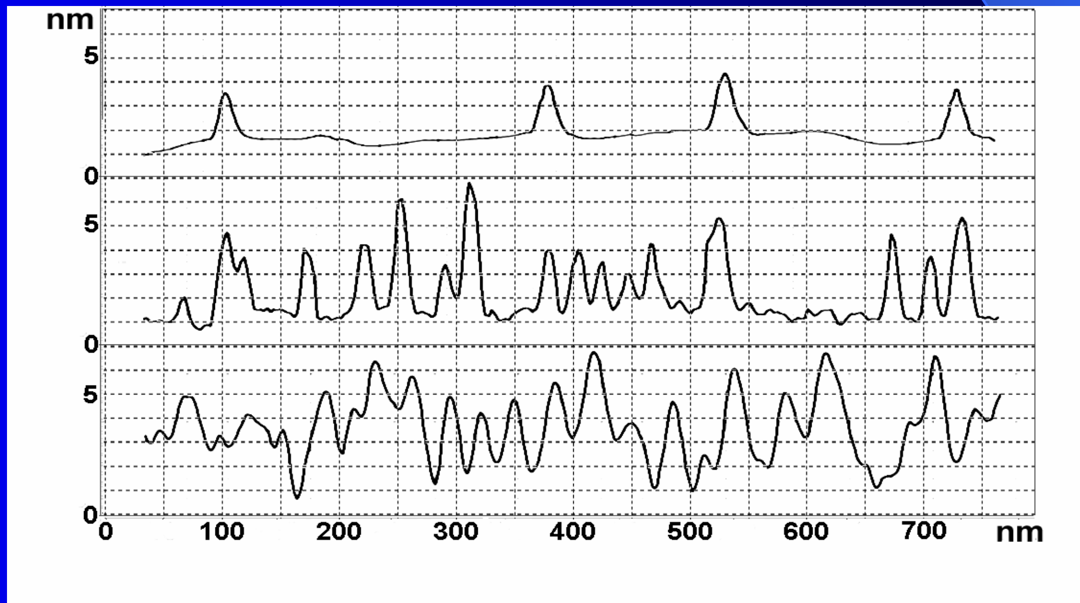


a b c

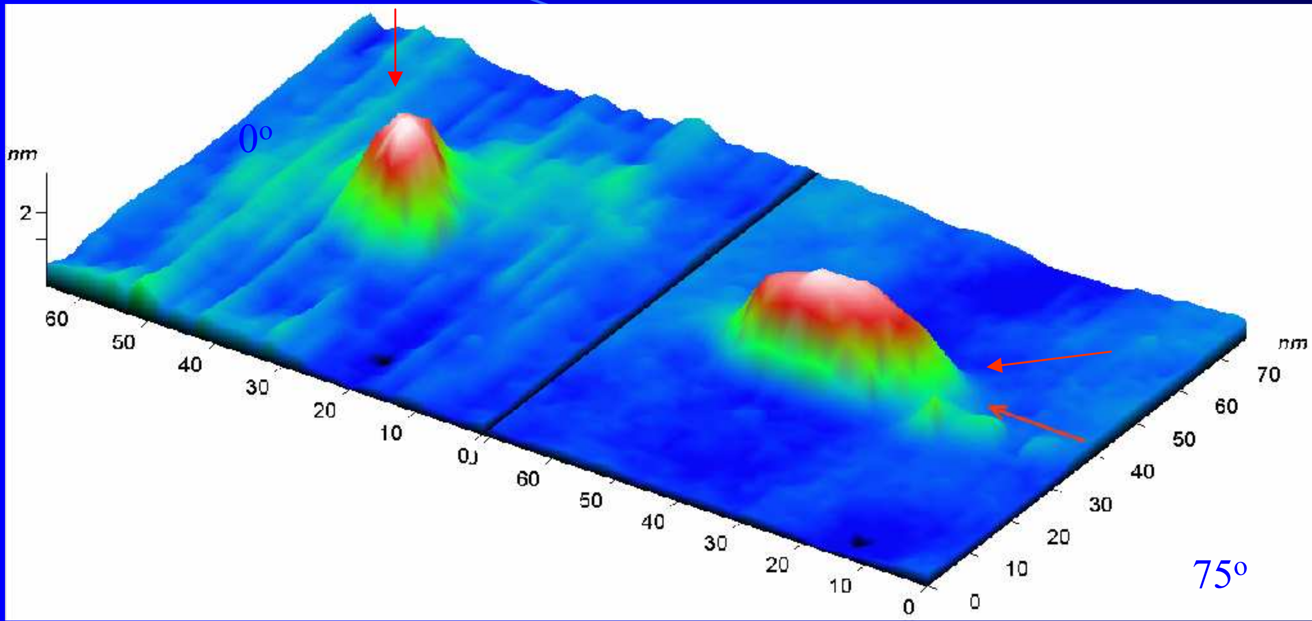
a - 2×10^{10} cm^{-2}

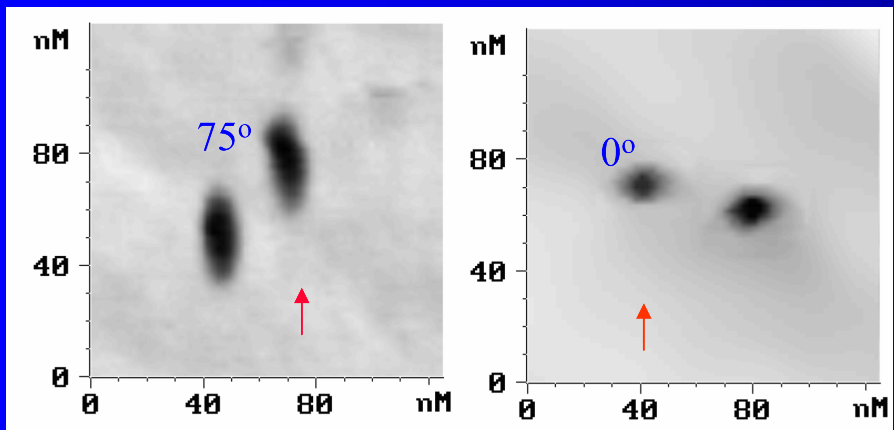
b- 1×10^{11} cm^{-2}

c - 1×10^{12} cm^{-2}

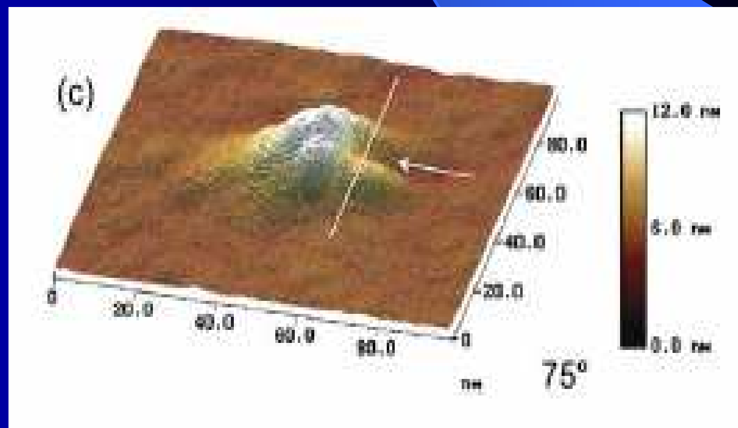


Variation of the hillock form on Al_2O_3 with Bi ion beam incidence angle





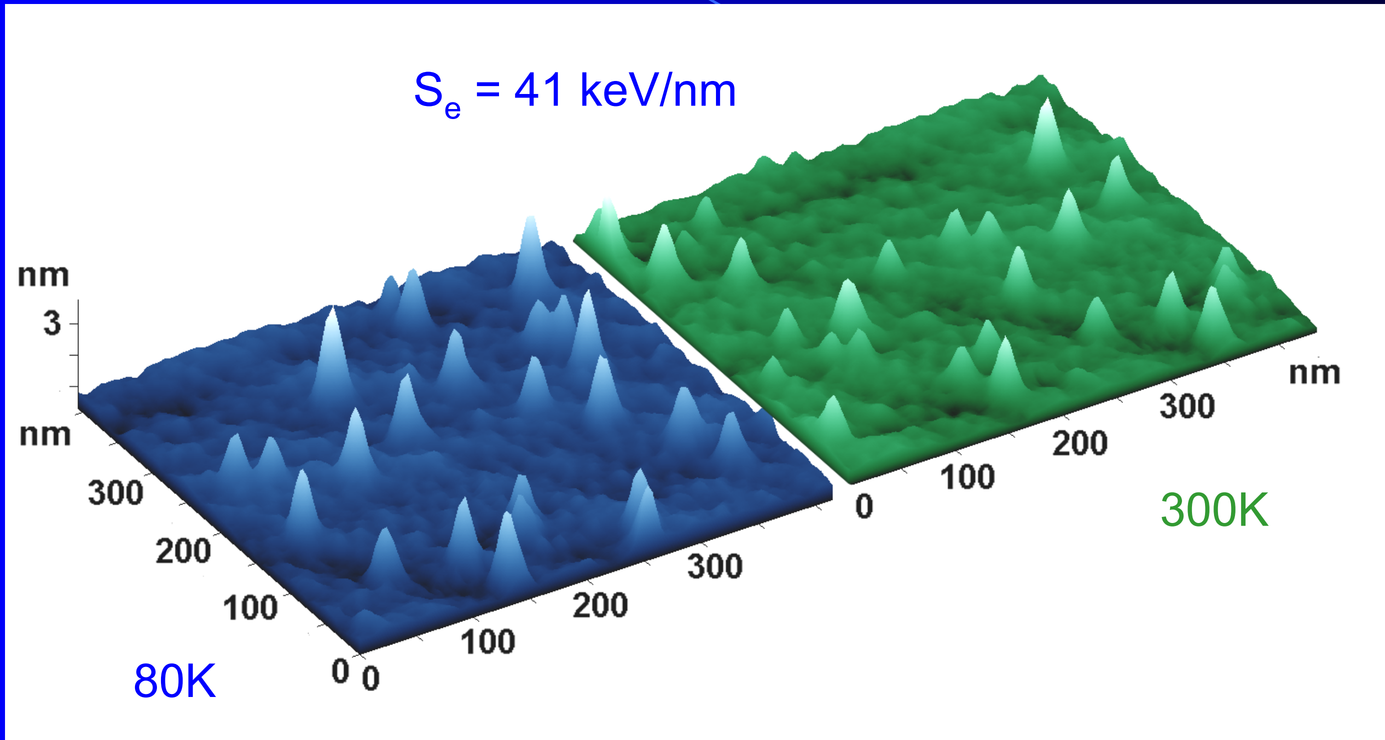
AFM phase images of α-A_2O_3 surfaces irradiated with 710 MeV Bi ions. Arrows show the ion beam direction



30 MeV C_{60} + $Fe_{0:55}Zr_{0:45}$ alloys

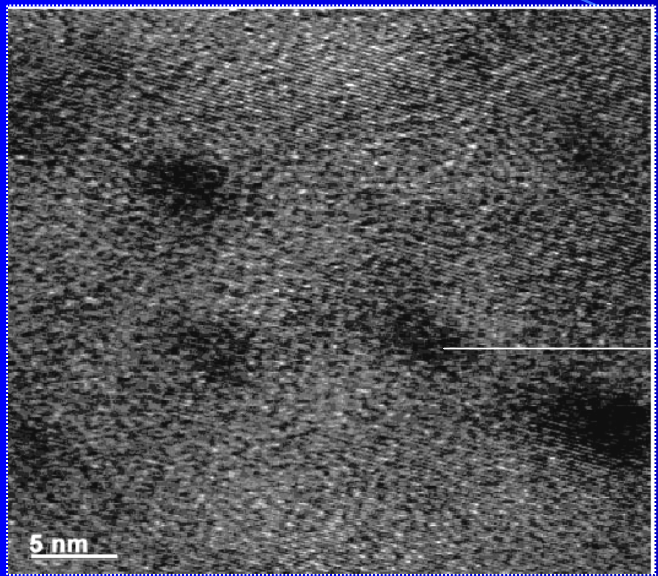
J.C. Girard et. al. Nucl. Instr. Meth., B 209 (2003) 85

3D AFM images of of α-A_2O_3 surfaces irradiated at 80K and 300K

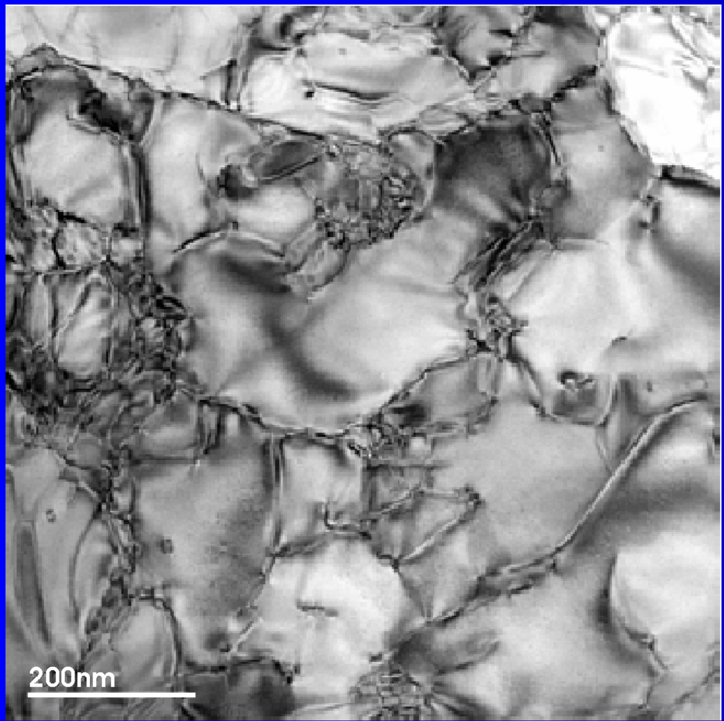


Thermal conductivity of sapphire: 1100 W/mK   (80 K)
30-40 W/mK (300 K)

TEM structural examination of sapphire at S_e=41 keV/nm



High-resolution lattice image of α-A_2O_3 irradiated with 710 MeV Bi ions a fluence of 7×10^{12} cm^{-2} at room temperature (plan-view specimen). The average TEM track diameter is ~3 to 4 nm.



TEM micrograph of α-A_2O_3 target irradiated at S_e=41 keV/nm to a fluence of 7×10^{12} cm^{-2} (S.J.Zinkle, ORNL)

The presence of numerous subgrains suggests that considerable internal stresses were induced by the Bi ion irradiation

The basic feature of surface defects formation is plastic deformation due to strain relaxation. The key questions are the nature of strain and the magnitude and dynamics of strain pulse in vicinity of the ion entrance point

<u>Temperature dependent mechanisms of the hillock-like damage formation</u>

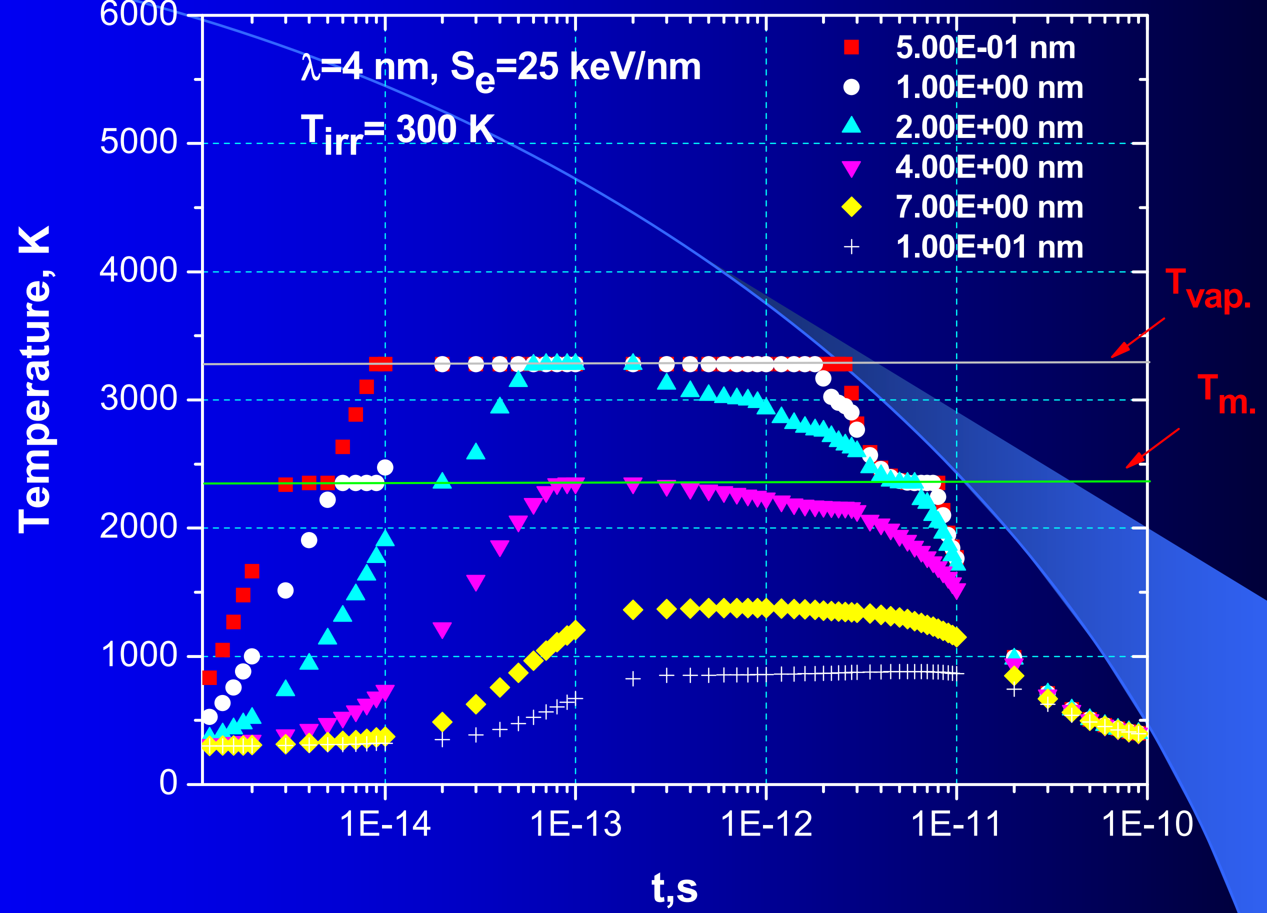
Hillock formation from the ion-induced melt due to the mechanical stresses as a result of thermal expansion
G. Szenes, Nucl. Instr. Meth., B 191 (2002) 31

mean hillock height $h \sim = gS_e + \beta S_n$

The necessary condition of hillocks production:

$T_p > 2.7T_0$, $T_0 = T_m - T_{irr}$. $2.7T_0 = 5535$ K

If $T_p < 2.7T_0$, cooling starts with shrinking in spite of the presence of the melt and no hillocks should be formed



Temperature evolution at different radial distances from the Bi ion axis in sapphire. T_m = 2340 K, vaporisation temperature is 3280 K. λ - electron-lattice interaction mean free path (calculated with TSPIKE02 code. *M. Toulemonde, C. Dufour, A. Meftah, E. Paumier, Nucl. Instr. and Meth., B 166–167 (2000) 903.*

The peak temperature, T_p < 4500 K, even on distance 0.5 nm from track axis, while $2.7T_0$ = 5535 K.

Temperature dependent mechanisms of the hillocks formation is not consistent with threshold electronic stopping power – melting temperature relation.

The hillocks are detected on MgO at lower energy deposition, although T_{melt} and $T_{vapor.}$ are higher than those known for Al_2O_3

Al_2O_3 $S_{thr.} \approx 25$ keV/nm $T_{melt.} = 2340$ K

MgO $S_{thr.} < 15.8$ keV/nm $T_{melt.} = 3245$ K

Hillock formation as a result of the Coulomb explosion due to charge imbalance in the subsurface region.

Quasineutrality of densely ionized region may be disturbed in the subsurface layer due to ejected electrons and incident ion charge neutralization process.

Baranov et al. Usp. Fiz. Nauk., 156 (1988), p. 477

Strong experimental indications for the occurrence of a macroscopic Coulomb explosion from a highly charged surface of crystalline Al_2O_3 under intense femtosecond laser pulse action have been reported recently

R. Stoian, D. Ashkenasi, A. Rosenfeld, and E. E. B. Campbell, Phys. Rev. B 62 (2000) 1367.

No surface profile modification have been detected under Kr^{+27} ion bombardment contrary to Bi^{+25} and Bi^{+17} ions. Ion charge neutralization cannot be the only condition for the hillock appearance.

Ion spike model allows to explain the absence of correlation between the arising of hillocks and formation of amorphous latent tracks in the bulk. Another indication in favor of Coulomb explosion is ellipsoidal form of the hillock baseline under tilted irradiation, reproducing the form of the spot with uncompensated charge on the surface and underlying region.

Summary

The structure of hillock-like defects on the surface of monocrystalline Al_2O_3, $MgAl_2O_4$ and MgO , induced by individual (0.5 ÷ 5) MeV/amu Kr, Xe and Bi ions, has been studied as a function of the ion energy, ion fluence, irradiation temperature and angle of ion incidence using scanning tunneling microscopy.

It was found that mean hillock height on sapphire surface depends linearly on the incident electron stopping power and increases in two times on average when hillocks start to overlap.

Noticeable changes in defect shape are registered only under strong deviation from normal beam incidence (more than 60 degrees) and no specific features (radial coherent mass transport outwards from the track core) typical for shockwave-like mechanism were observed.

The hillocks on the surface of α-Al_2O_3 are observed at ionizing energy loss less than threshold value of the phase transformation and new mechanisms for explanation of individual surface damage production are required.

As a possible reason of hillocks formation, the plastic deformation due to the defects created by the Coulomb explosion mechanism in the target subsurface layer is suggested.

Plans for the future

I. Determination of threshold electronic stopping power values for nanoscale defects formation for new ion + target combinations. Target materials to be involved in experiments: *ZrO_2, ZrC, TiC, AlN.*

II. High-resolution transmission electron microscopy studies of ceramic materials irradiated with heavy ions of fission fragment energy to elucidate the correlation between surface and material bulk radiation damages.

III. Elaboration of atomostic mechanisms responsible for the hillocks production on base MD simulation methods.