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Synthesis, Modification and Characterization"

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Phase Transitions in Pressureized Solids Triggered by Energetic Heavy Ions

Maik Lang
GSI - Materials Research Department
Darmstadt, Germany

Strada Costiera 11, 34014 Trieste, Italy - Tel. +39 040 2240 111; Fax +39 040 224 163 - sci_info@ictp.it, www.ictp.it

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Maik Lang

GSI - Materials Research Department, Darmstadt (Germany)

Collaboration of Physicists and Geoscientists

U.A. Glasmacher	MPI & Univ. Heidelberg
G.A. Wagner	MPI Heidelberg
H. Keppler	Univ. Bayreuth (BGI)
F. Langenhorst	Univ. Jena
M. Lang R. Neumann D. Schardt C. Trautmann	GSI – Darmstadt

Motivation

Radioactive decay products (fission fragments, alphas, recoils)
in the interior of the Earth

ion-matter interaction

★ heat source → differentiation of Earth

Press & Siever: *Earth*, WHF (1986).

★ source for power geodynamo today

Lee & Jeanloz, Geophys. Res. Lett. **30** (2003) 2212.

★ basis for dating in geochronology

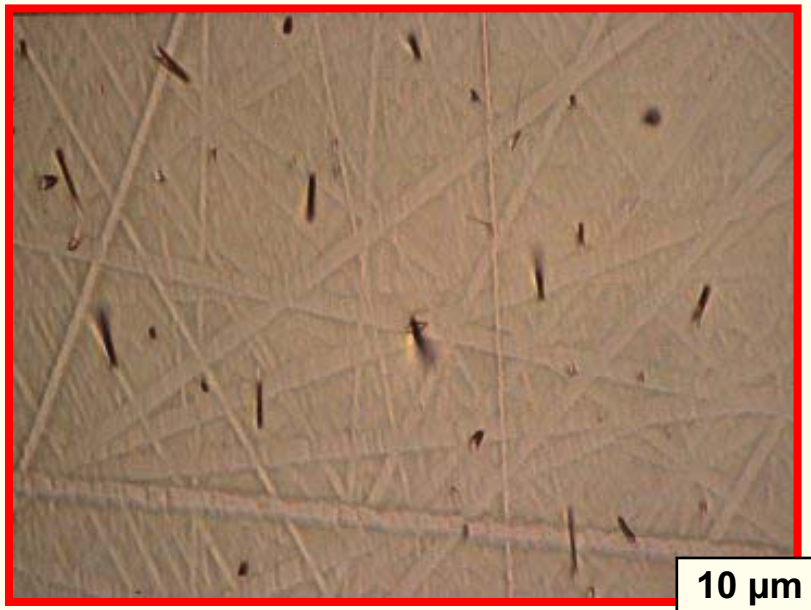
Wagner et al., Ferdinand Enke Verlag,1992

Fission Tracks

daughter nucleus 1 daughter nucleus 2

not annealed

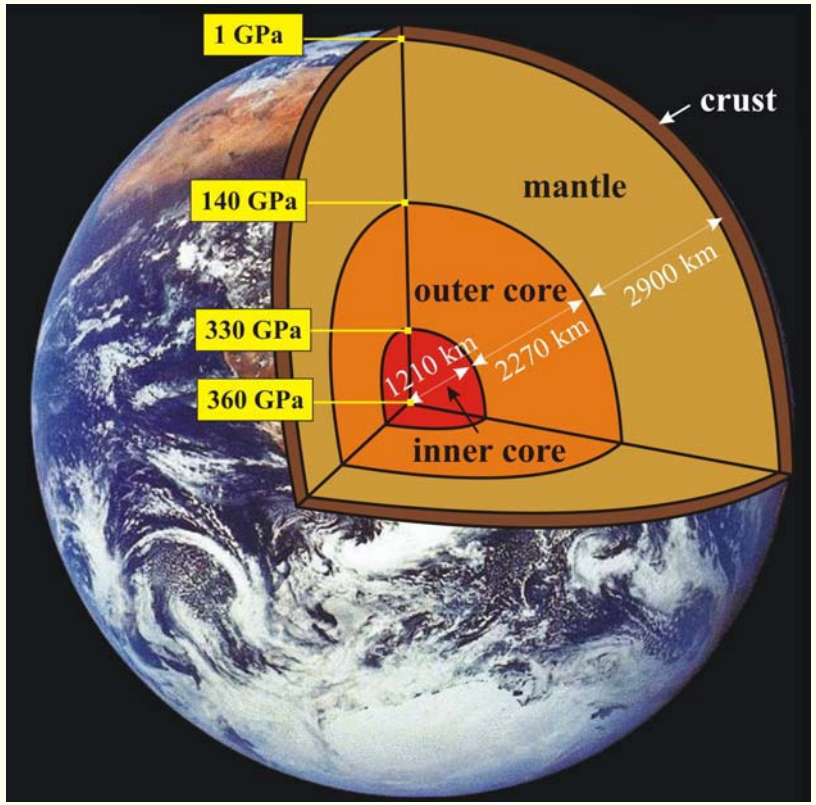
annealed (350 °C, 1 hour)



(www.geotrack.com.au)

Motivation

Minerals exposed to high pressures and temperatures



★ influence of pressure on ion-track formation?

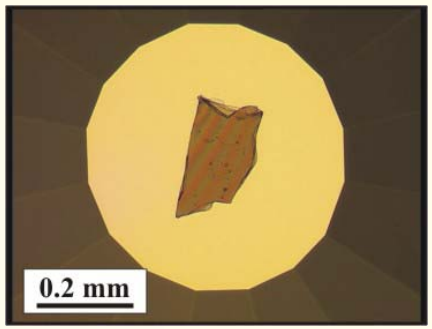
(e.g. track length → dating)

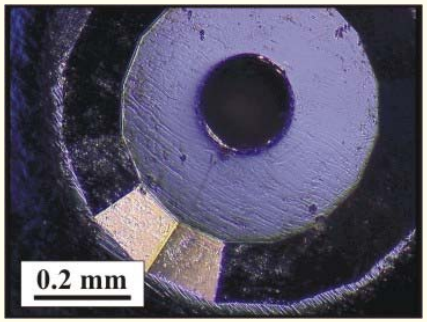
★ can ions induce specific phase transition in pressurized solids?

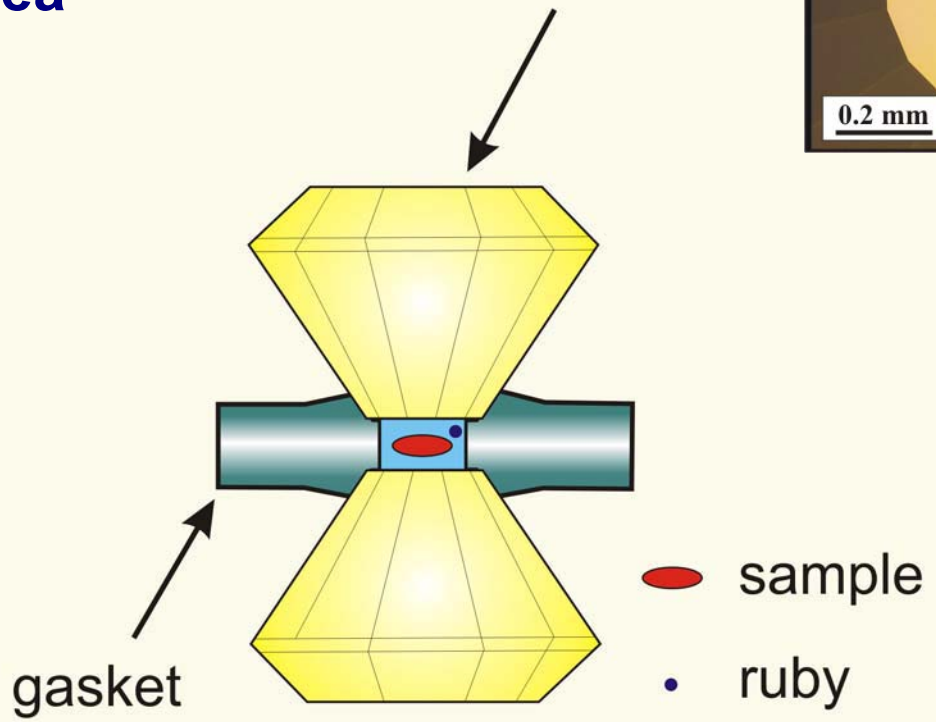
➡ Irradiation of pressurized solids at GSI

Experimental setup: DAC

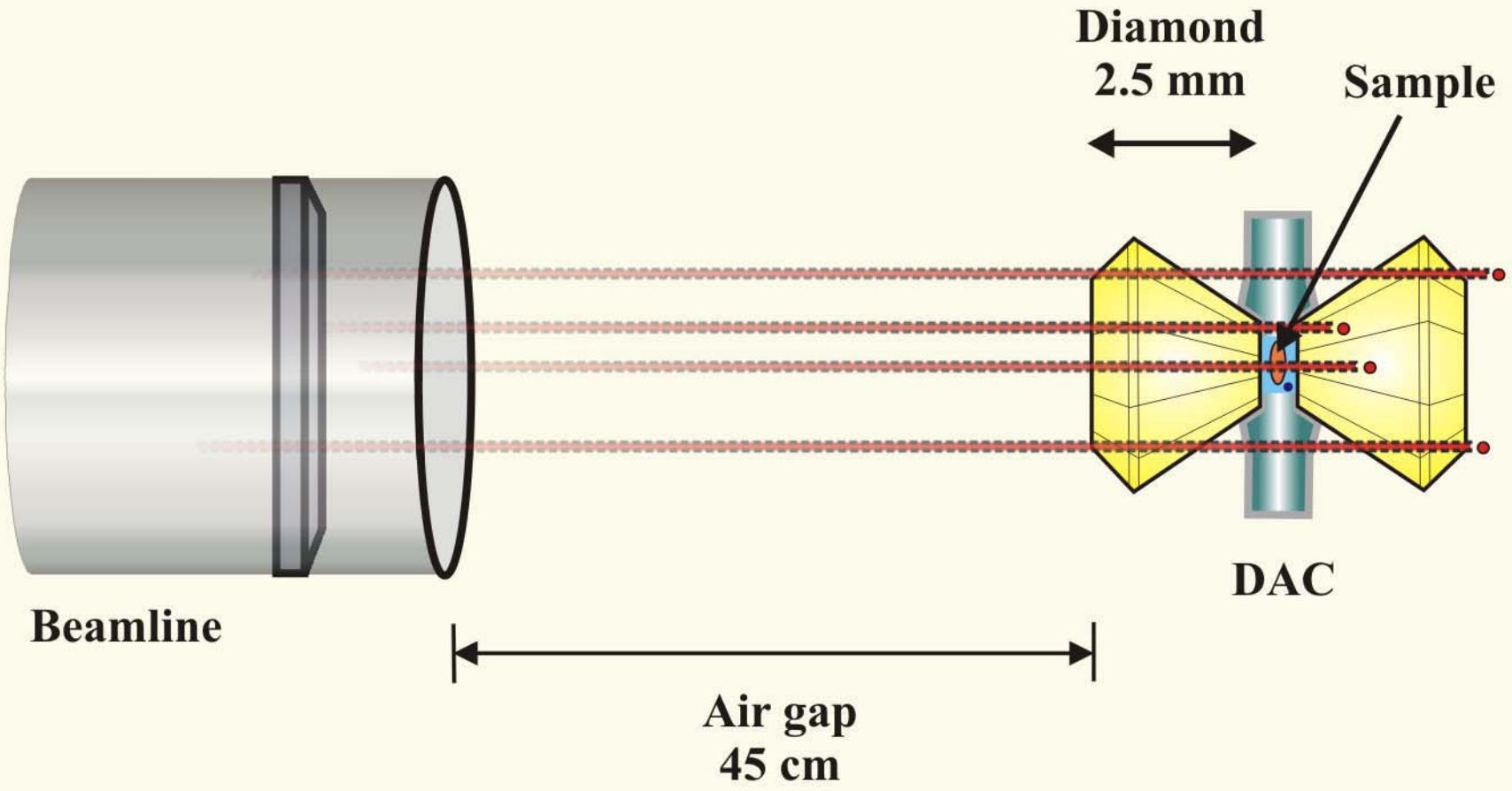
$$\text{pressure} = \frac{\text{force}}{\text{area}}$$

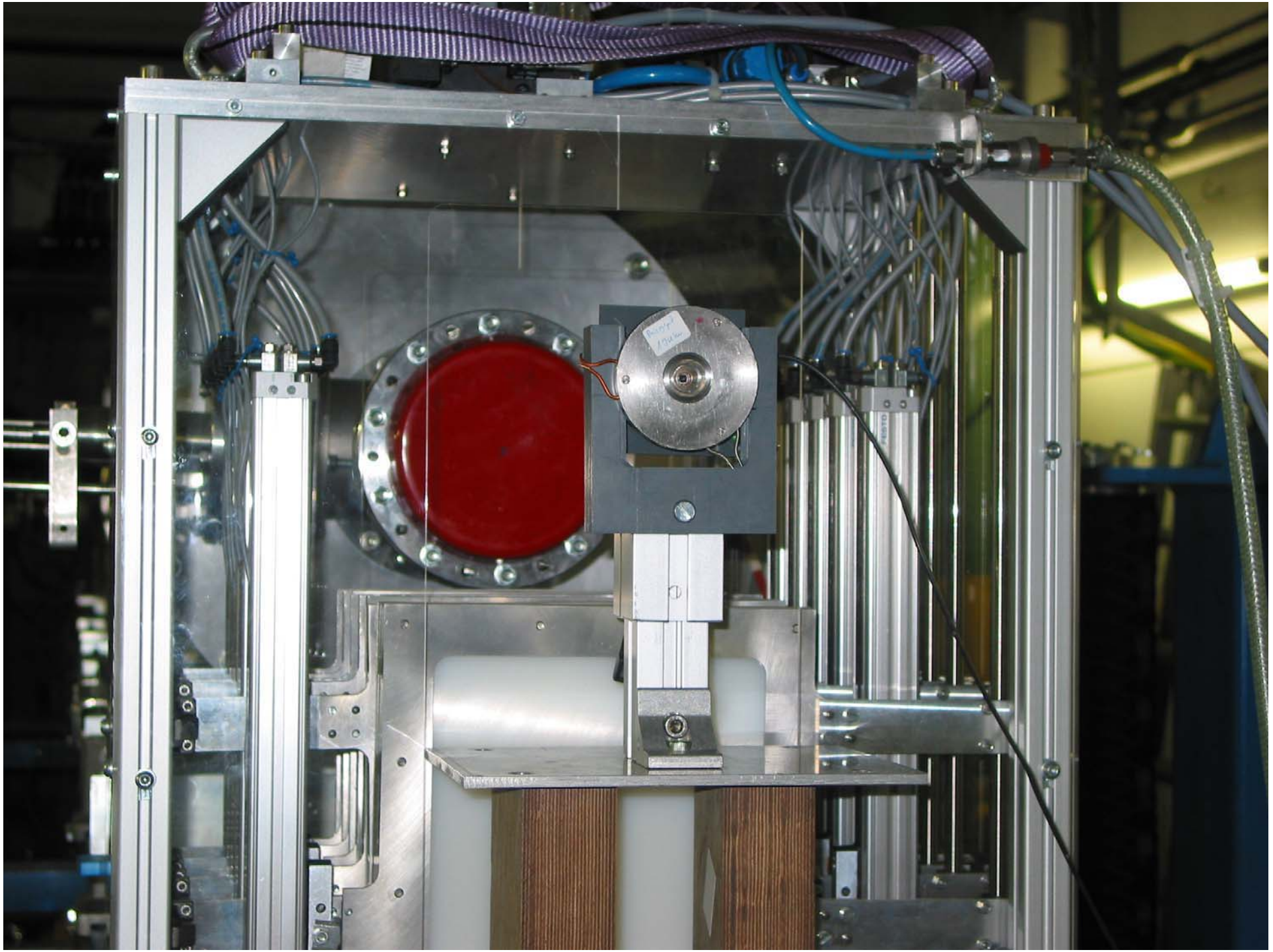




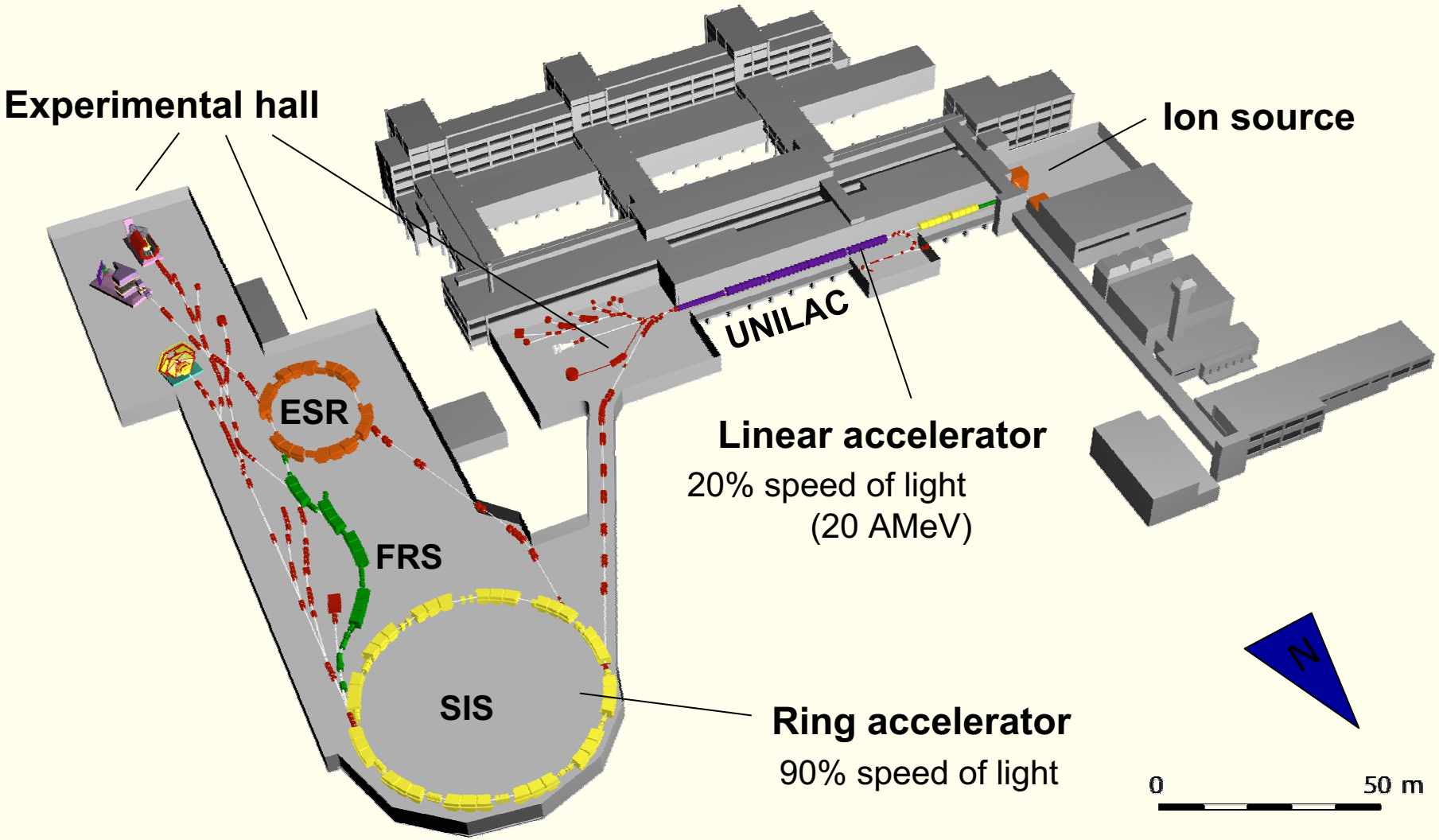


Experimental setup

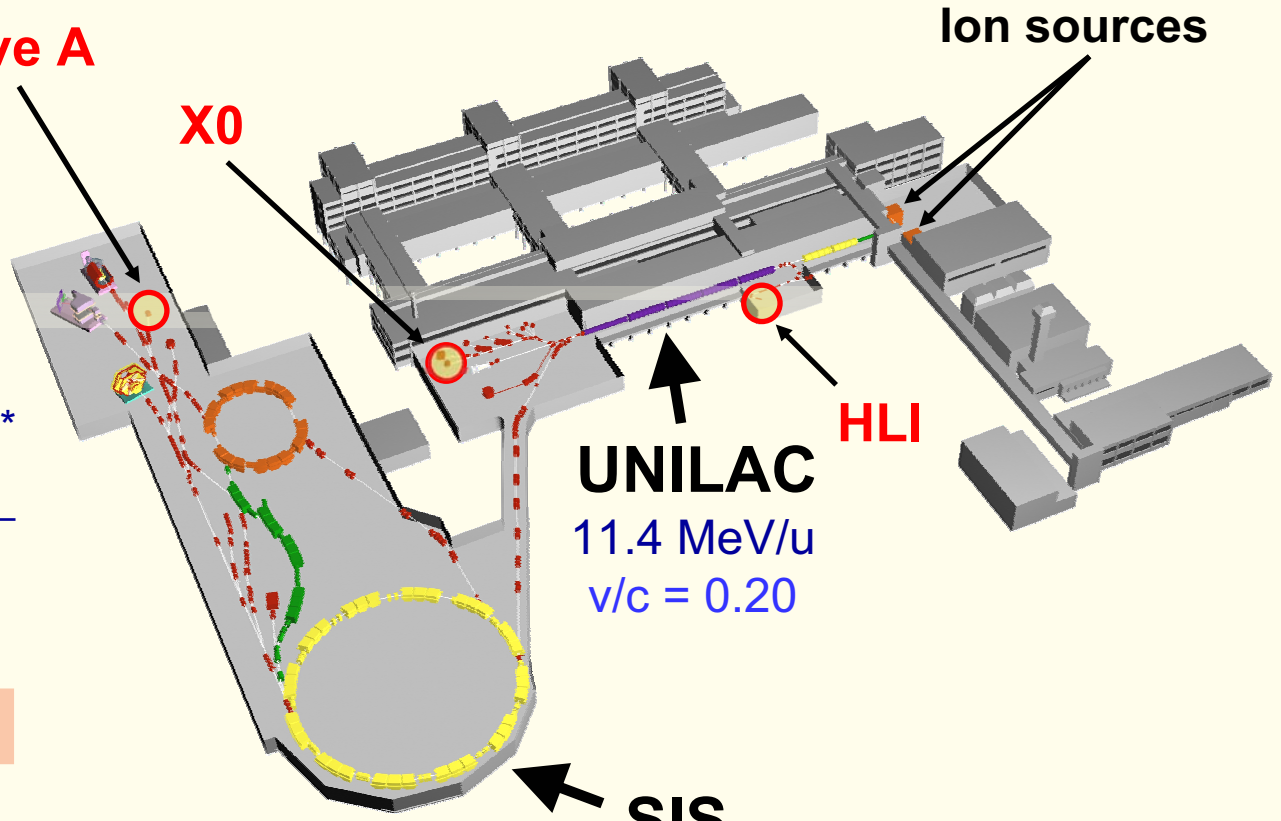




GSI heavy-ion accelerators



Experimental setup: Irradiation facility



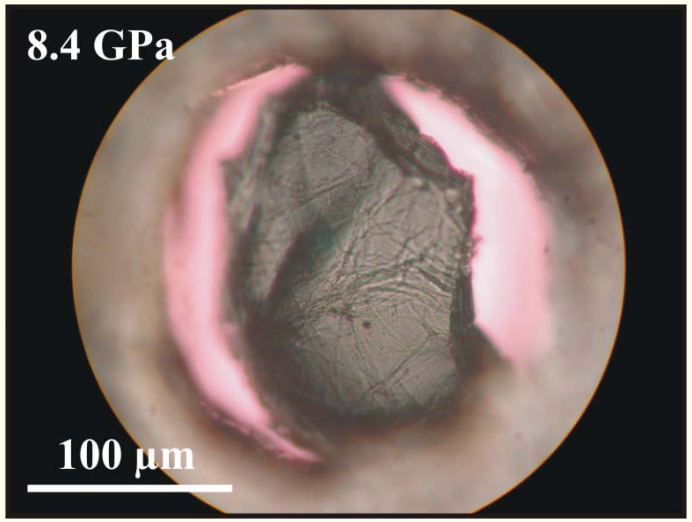
Energy [MeV/u]	Energy [GeV]	Range* [mm]
1.4	0.3	0.001
11.4	2.7	0.07
200	47.6	2.7
500	119	11.6
1000	238	30.0

*Range of ^{238}U in diamond, calculated with SRIM03

Samples

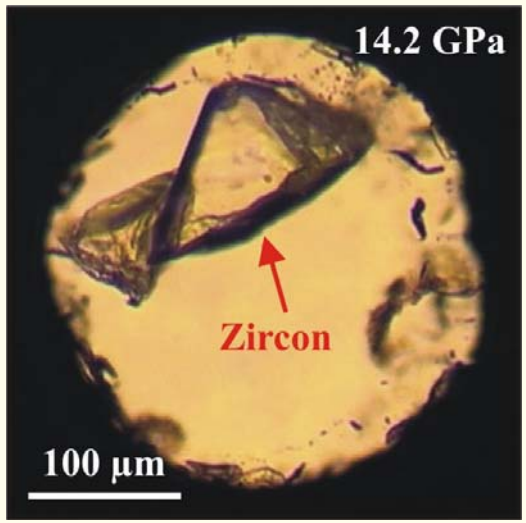
Graphite

HOPG (ρ = 2.27 g/cm^3)



Zircon

nat. $ZrSiO_4$ (ρ = 4.70 g/cm^3)



pressure during irradiations

P = 1 bar

P = 0.5 GPa

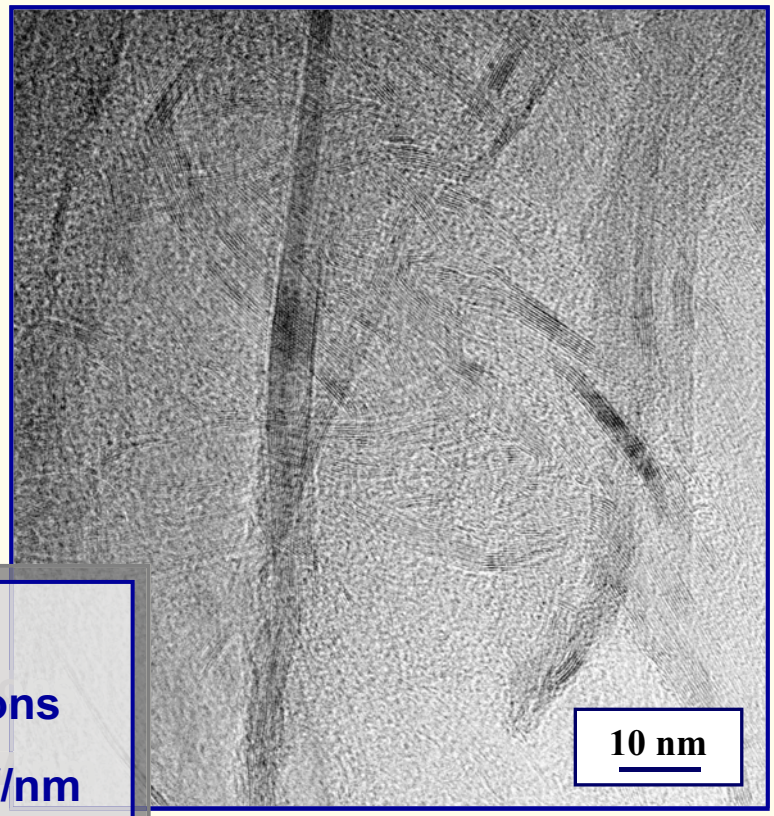
P = 8.4 GPa

P = 12.1 GPa

P = 14.2 GPa

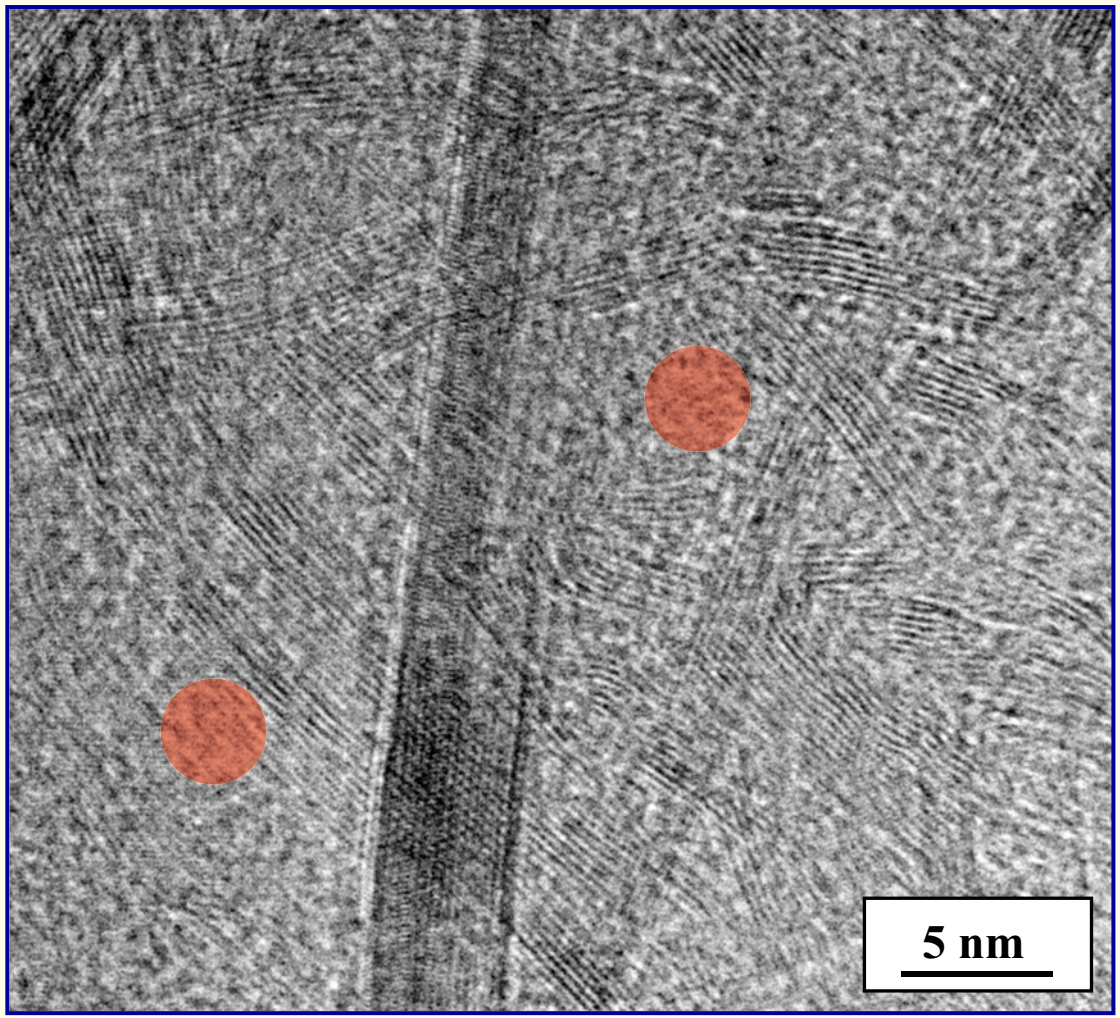
Results: Graphite

Transmission electron microscopy



- P = 8.4 GPa
- 40.5 GeV ^{238}U ions
- dE/dx = 8.0 keV/nm
- 1×10^{11} ions/cm^2

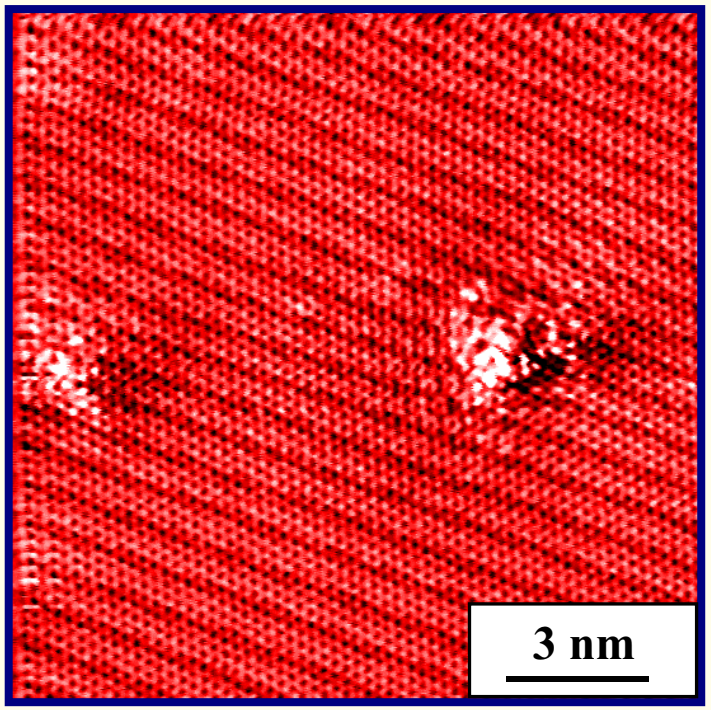
Results: Graphite



Results: Graphite

Ambient pressure sample:

- P = 1 bar
- 2.6 GeV ^{238}U ions
- dE/dx = 27 keV/nm
- $1 \cdot 10^{11}$ ions/cm^2



Scanning tunneling microscopy:

- amorphous tracks in crystalline matrix
- size of tracks ~3 nm
- number of tracks ↔ ion fluence

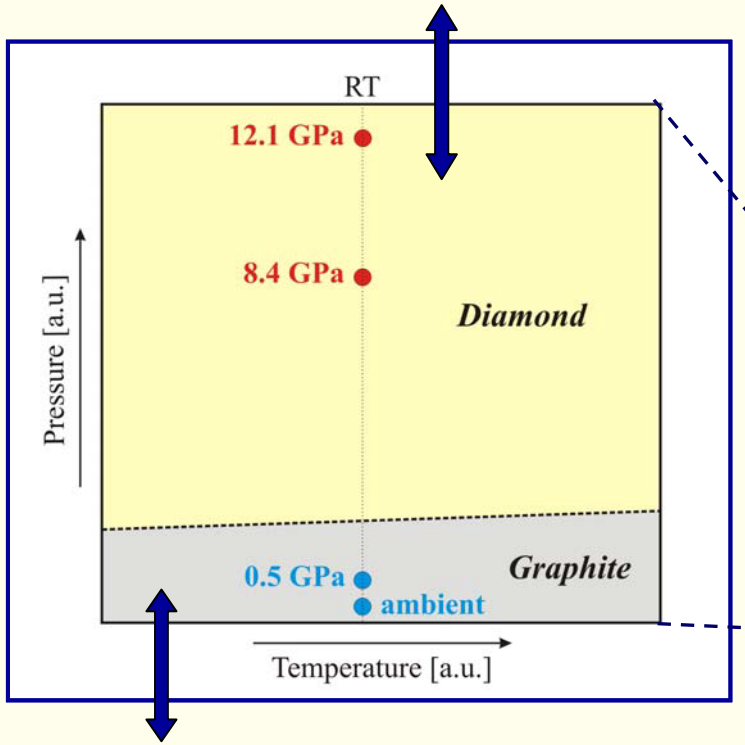
➡ consistent with earlier investigations ✓

(J. Liu et al., Phys. Rev. B **64** (2001) 184115.)

Results: Graphite

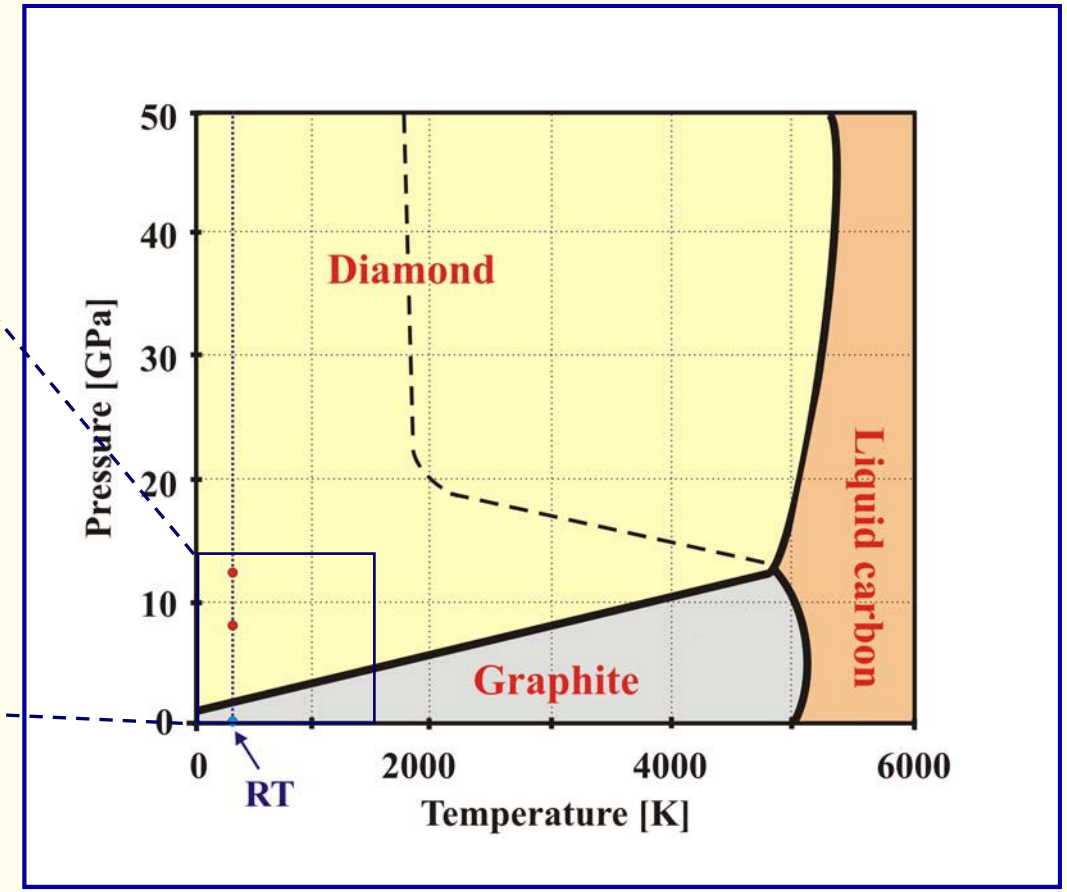
outside stability field of graphite

amorphous graphite +
crystalline bands



inside stability field of graphite

graphite remains crystalline
no bands



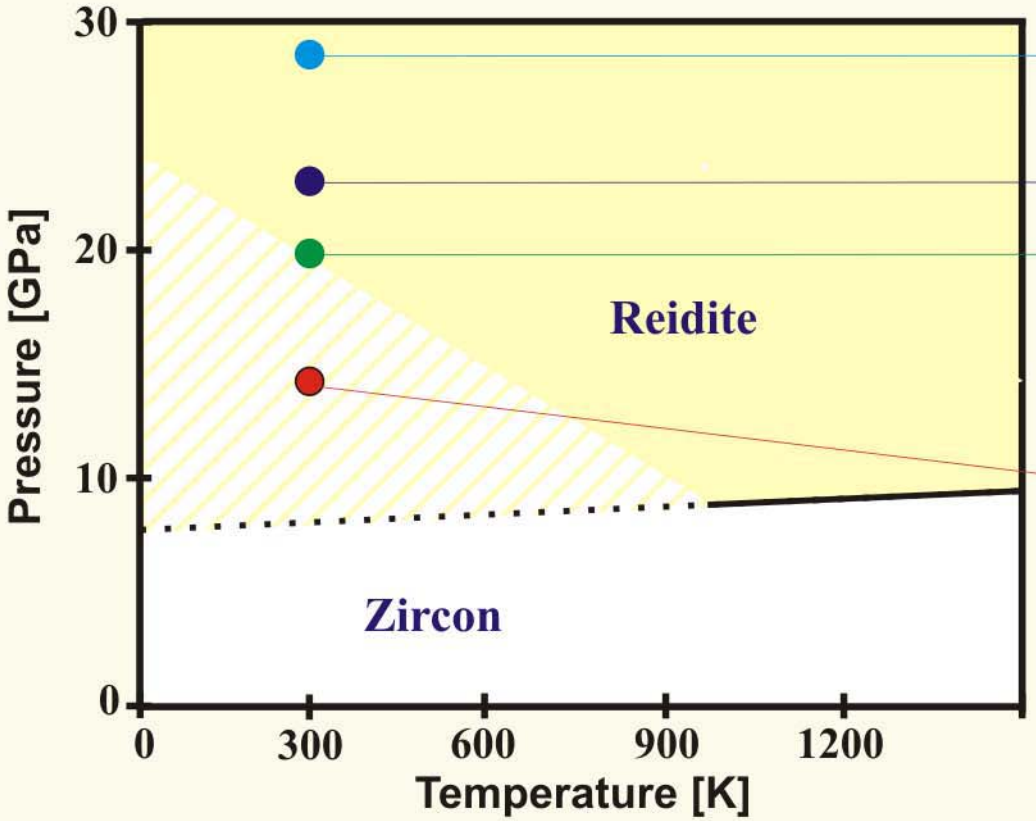
Zircon

Zircon at high-pressure conditions:

- high-pressure phase reidite
  (1969 by Alan Reid, high-pressure experiment)
- reidite discovered in nature 2001
  (meteorite impact region)
- reidite 10% denser than zircon and quenchable to ambient pressure

→ Critical pressure of zircon-reidite transition?

Results: Zircon



shock experiment (nat. zircon)

DAC experiment (nat. zircon)

DAC experiment (synth. zircon)

DAC-irradiation experiment (nat. zircon)

14 GPa !

Results: Zircon

Raman spectroscopy

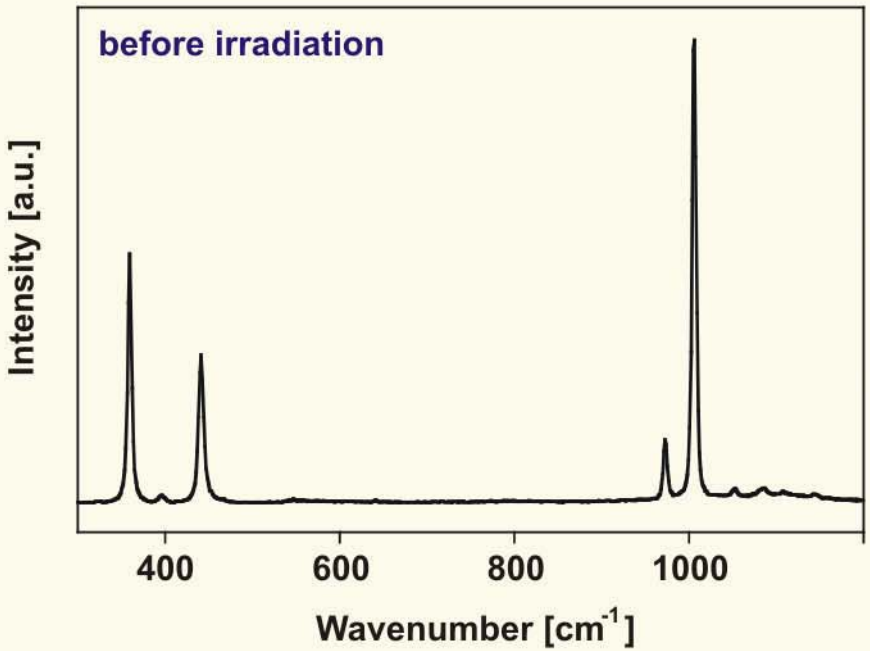
^{238}U ions

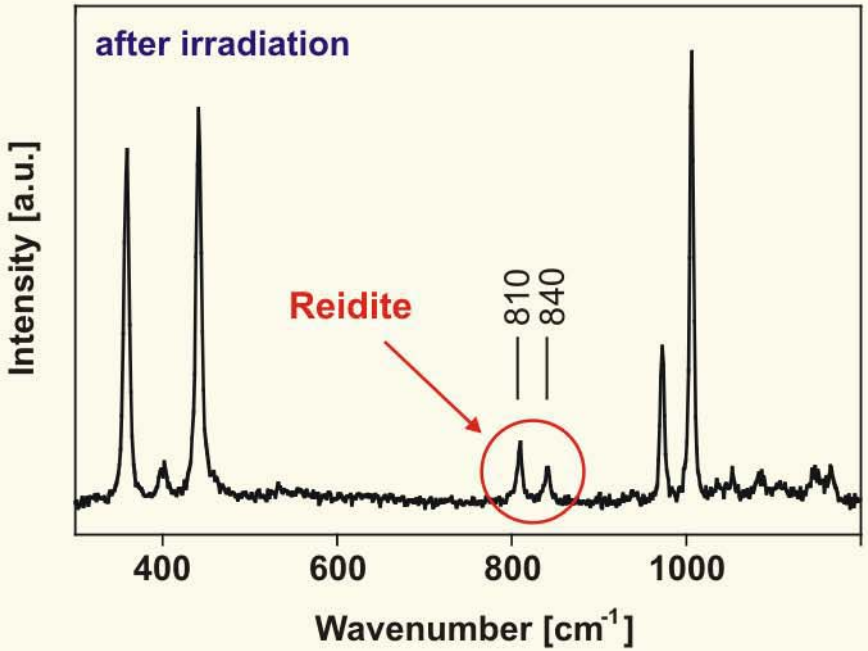
P = 14.2 GPa

E = 13.8 GeV

dE/dx = 24.5 keV/nm

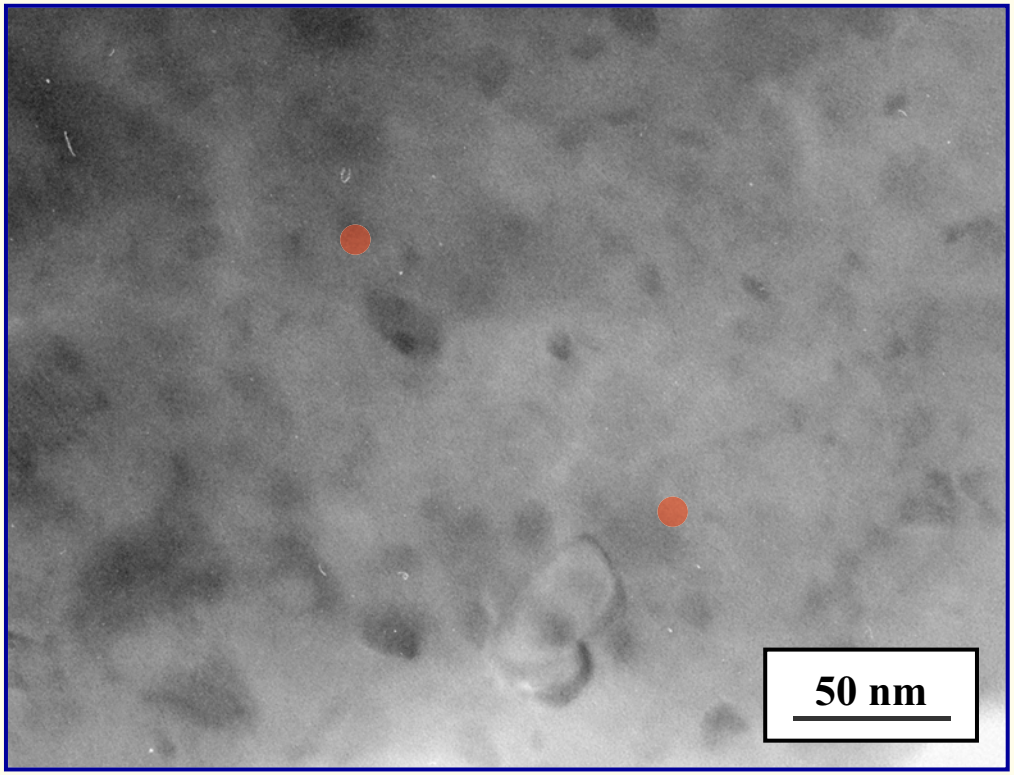
2×10^9 ions/cm^2

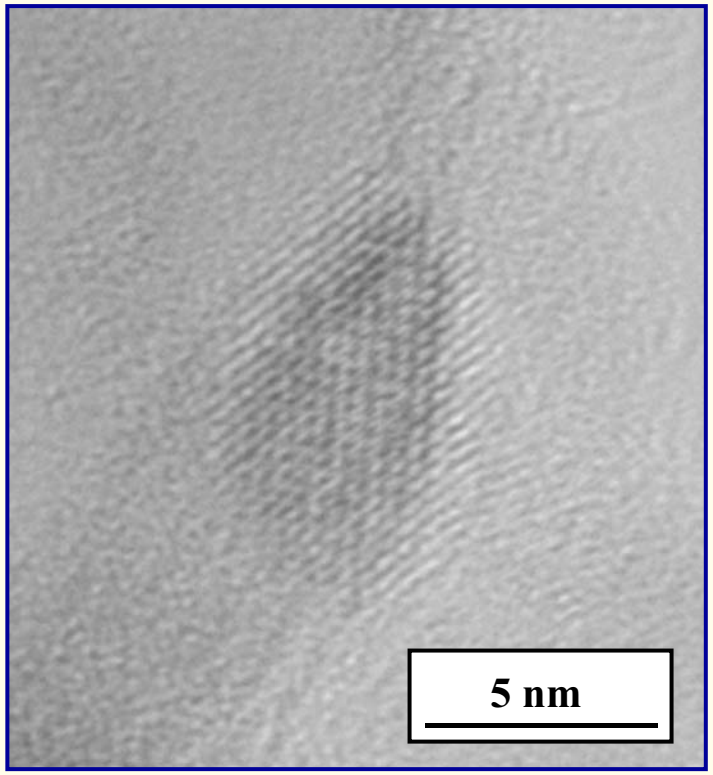




Results: Zircon

Transmission electron microscopy





Results: Zircon

previous high-pressure experiments (RT)

P = ambient to 20 GPa:

- no zircon-reidite transition
- no fragmentation

(Van Westeren et al., Am. Mineral. **89** (2004) 197)

previous ion-irradiation experiments

P = ambient:

- amorphous tracks (size 8 nm) in crystalline zircon matrix

(Bursill & Braunshausen, Philos. Mag A **62** (1990) 395)

high pressure + irradiation

P = 14.2 GPa

- reidite nanocrystals
- fragmentation
- no ion tracks

(Glasmacher et al., Phys. Rev. Lett. **96** (2006) 195701)

Results: Zircon

Reidite formation at pressures below critical value (20 GPa)

➡ **possible reasons:**

ions induce local heating → lowering of critical pressure

(Bursill & Braunshausen, Philos. Mag A **62** (1990) 395)

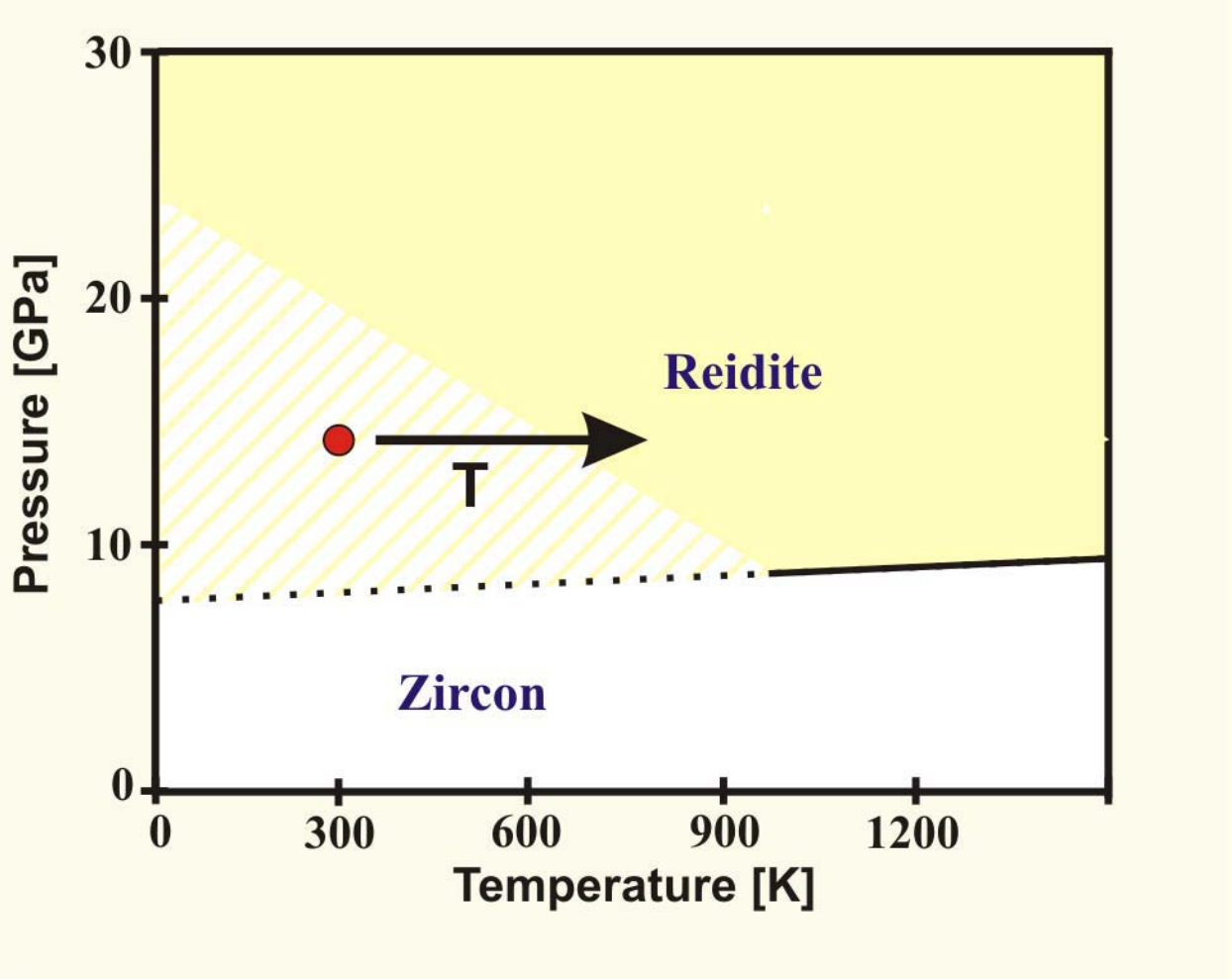
(Ono et al., Am. Mineral. **89** (2004) 185)

ions induce additional pressure → increase of effective pressure

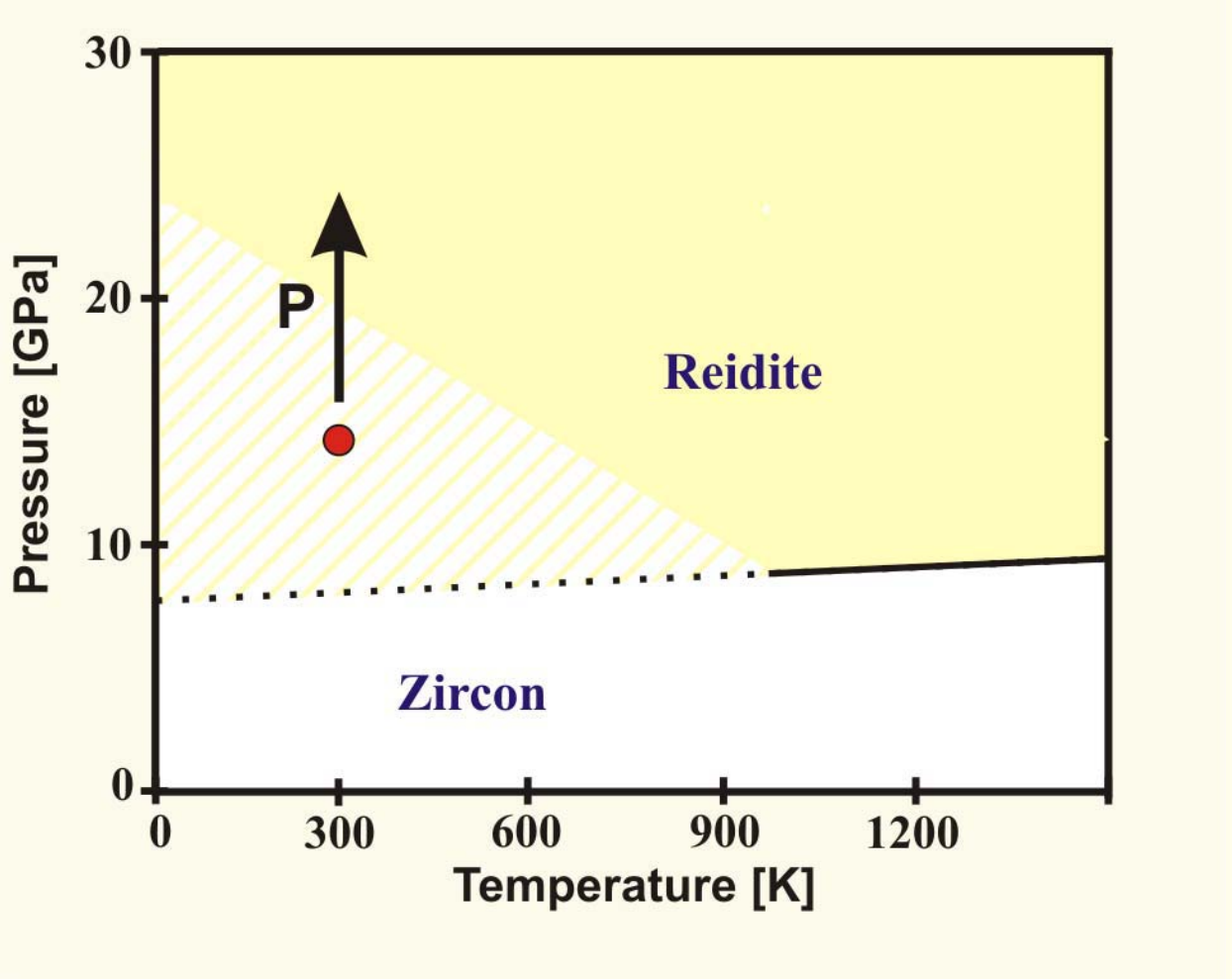
(A. Gucsik et al., Mineral. Mag. **68** (2004) 801)

ions induce defects → lowering of activation energy

Results: Zircon



Results: Zircon



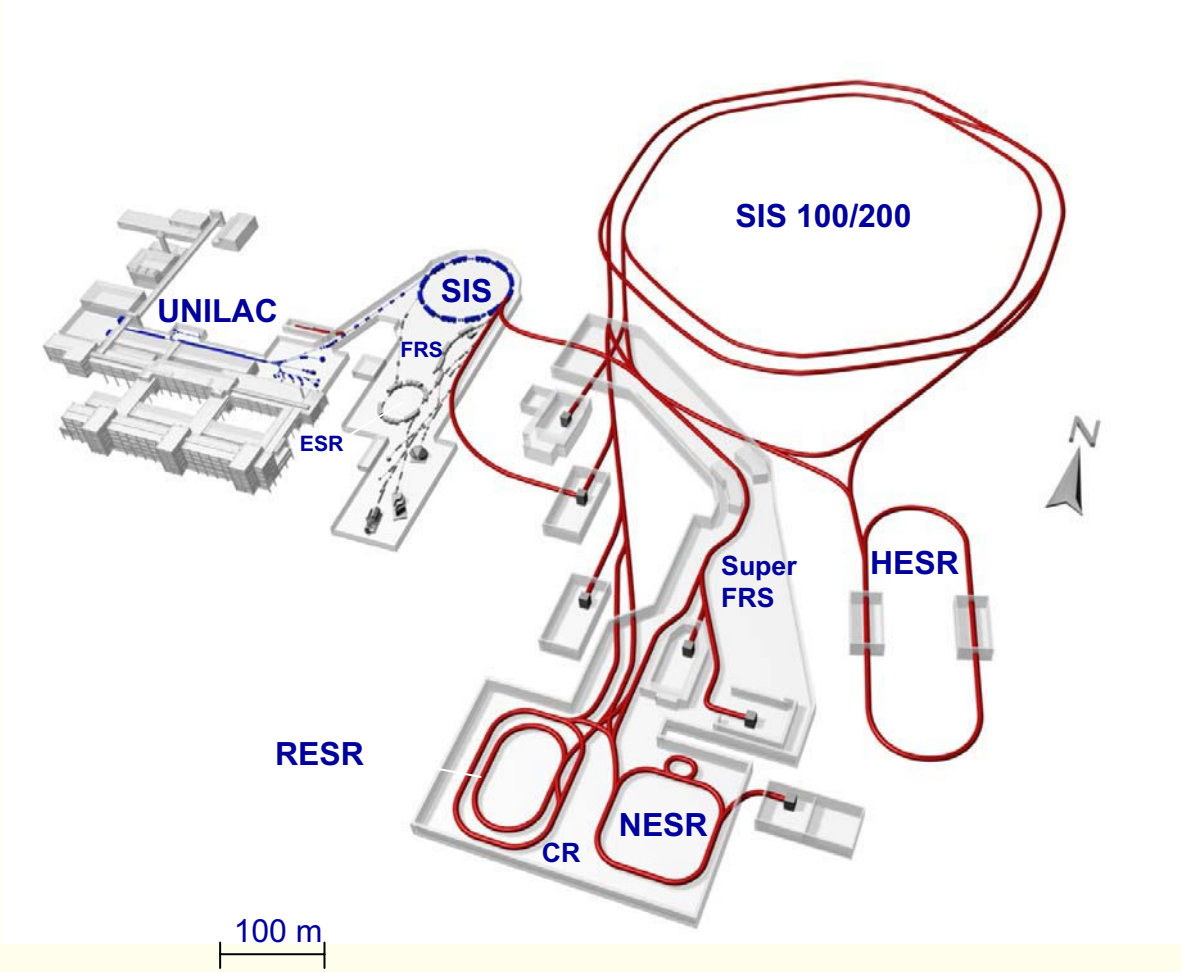
Conclusions

- Ion irradiation of solids in pressure cells demonstrated up to 22 GPa (220 kbar)
- Ion-induced modifications of graphite depend on pressure
- Ion-induced displacive phase transition in zircon at pressure well below critical value
- Pervasive changes in microstructure of solids (fragmentation, crystallization)

Outlook

- Irradiations of pressurized samples at high temperature
- Irradiation of zircon at realistic crust and mantle conditions
  (up to 5 w% ^{238}U & $^{232}Th \rightarrow 10^7$ fission-tracks/cm^2 and 10^{14} α's/cm^2 (100 Ma))
- Extension of high-pressure irradiations to other minerals
  (e.g. olivine)
- Irradiations of materials at extreme pressure conditions
- Extension to larger sample volumes

GSI – Future Facility



New beams

Exotic nuclei

Antiprotons

New beam qualities

Beam intensity

Beam precision

GSI – Future Facility