# Modelling high-energy radiation damage in nuclear power and fusion applications

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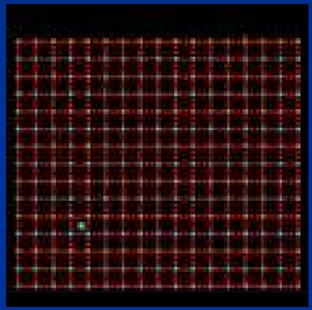
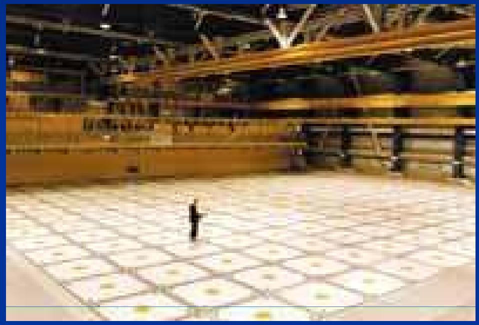
# Radiation damage effects in ceramics

- Motivation: pressing need to safely encapsulate radioactive nuclear waste that comes from power plants and surplus plutonium





- HLW accounts for 95% of the total radioactivity produced during nuclear electricity generation. The amount of HLW worldwide is increasing by 12,000 metric tons a year, equivalent to 100 double-decker buses

- Pu stockpile in the UK: ~100 metric tons (reprocessed from waste and from decommissioned weapons), of which 20 tons has been declared as waste (Am)

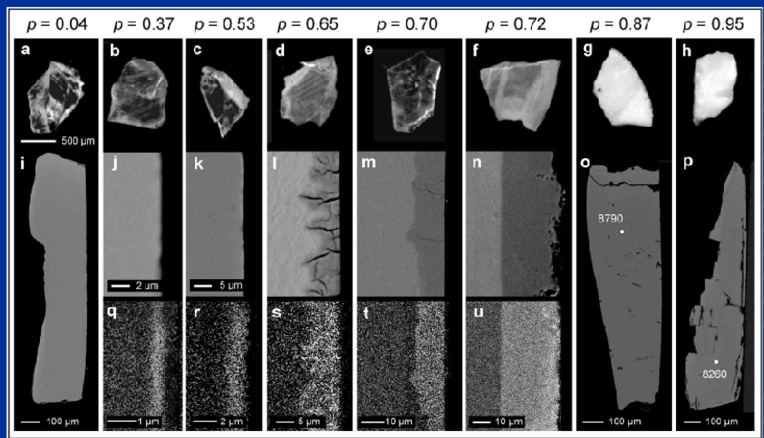
- Ceramics are proposed to be used for encapsulation of high-level waste

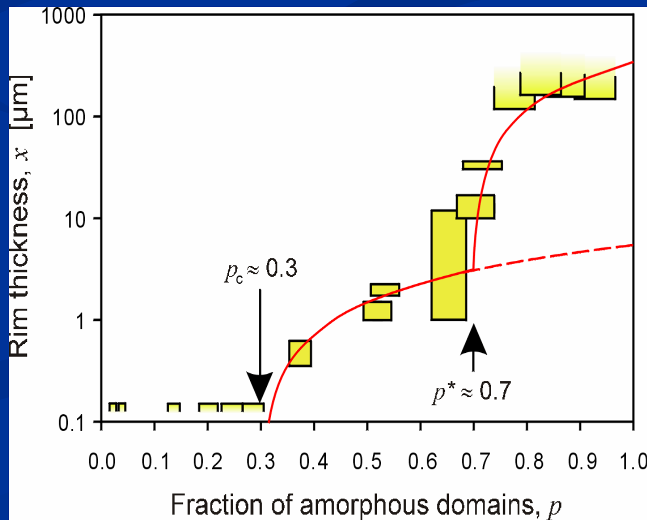
# Effect of radiation-induced amorphization on diffusion

Actinides are long-lived. A waste form should be stable during millions of years. Traditional glasses are not an option. UK NDA and NNL want to use ceramics.

Case study: zircon $ZrSiO_4$ found minerals are ~1 billion years old, completely amorphous yet intact

Absorbs large ions like Pu on Zr site

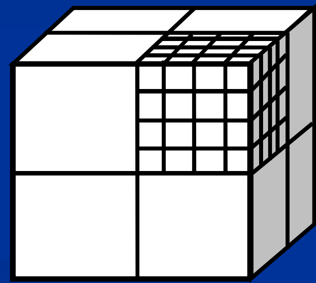


## Molecular dynamics simulation of radiation damage

**Details:**

1. Empirical potentials and short-range ZBL potential at short <~1 Å distances

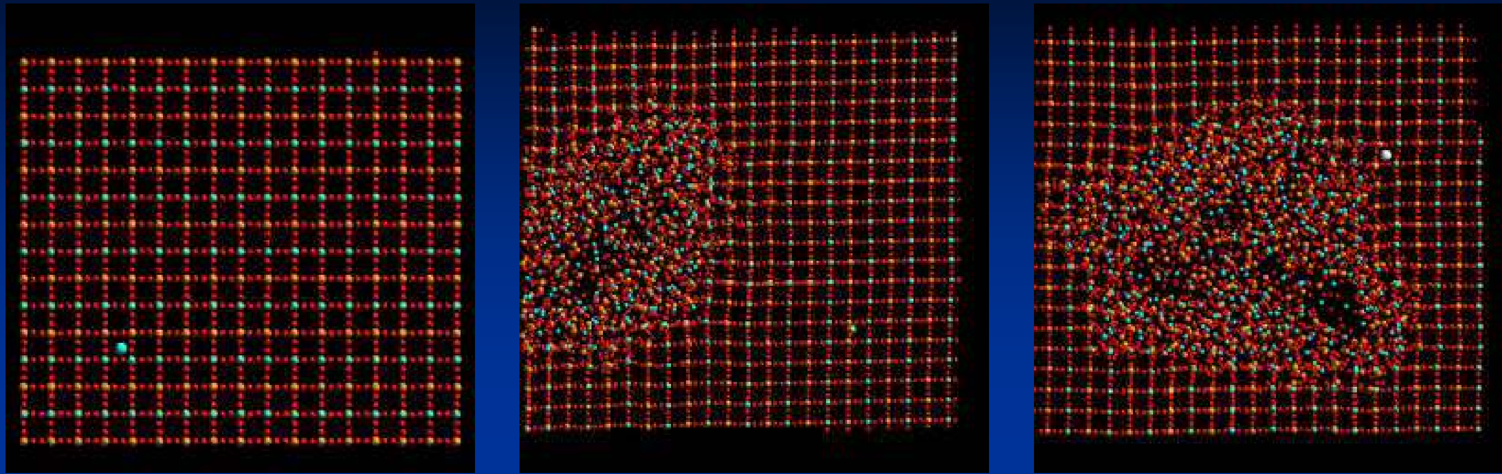


2. Almost **perfectly scalable** MD code based on domain decomposition strategy (DL_POLY 3, 4)

3. Parallel computers
Cambridge HPC, HPCx, HECToR
(time through Materials Chemistry Consortium)

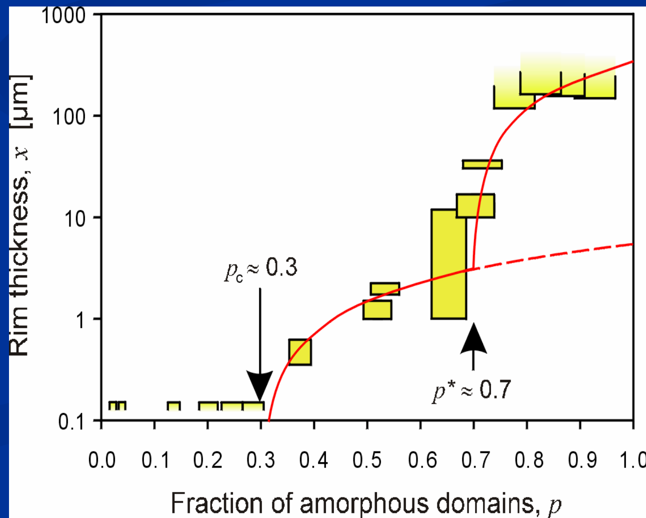
4. Adapted MD code to handle out-of-equilibrium conditions (variable time step, boundary scaling, inhomogeneous density) and to analyze radiation damage on the fly

70 keV
U recoil



Channels of low density appearing along the track

This gives channels of increased diffusion and explains percolation-type increases of transport at $p$=0.3



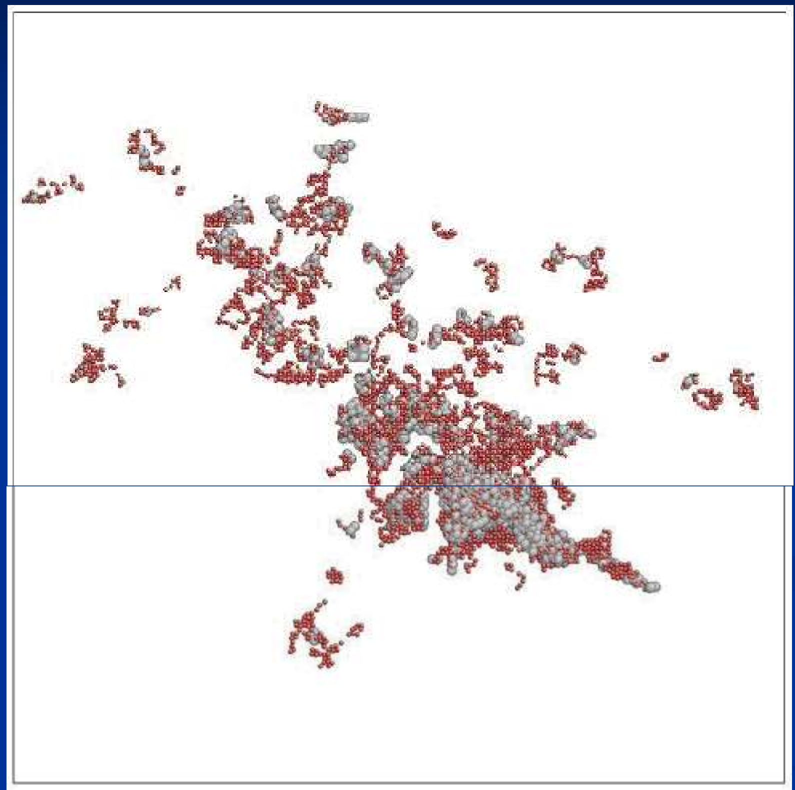
# Resistant vs amorphizable materials

$Gd_2Ti_2O_7$ – “official” US Department of Energy waste form.
Amorphizes easily under irradiation

$Gd_2Zr_2O_7$ – does not amorphize even at very large radiation doses!

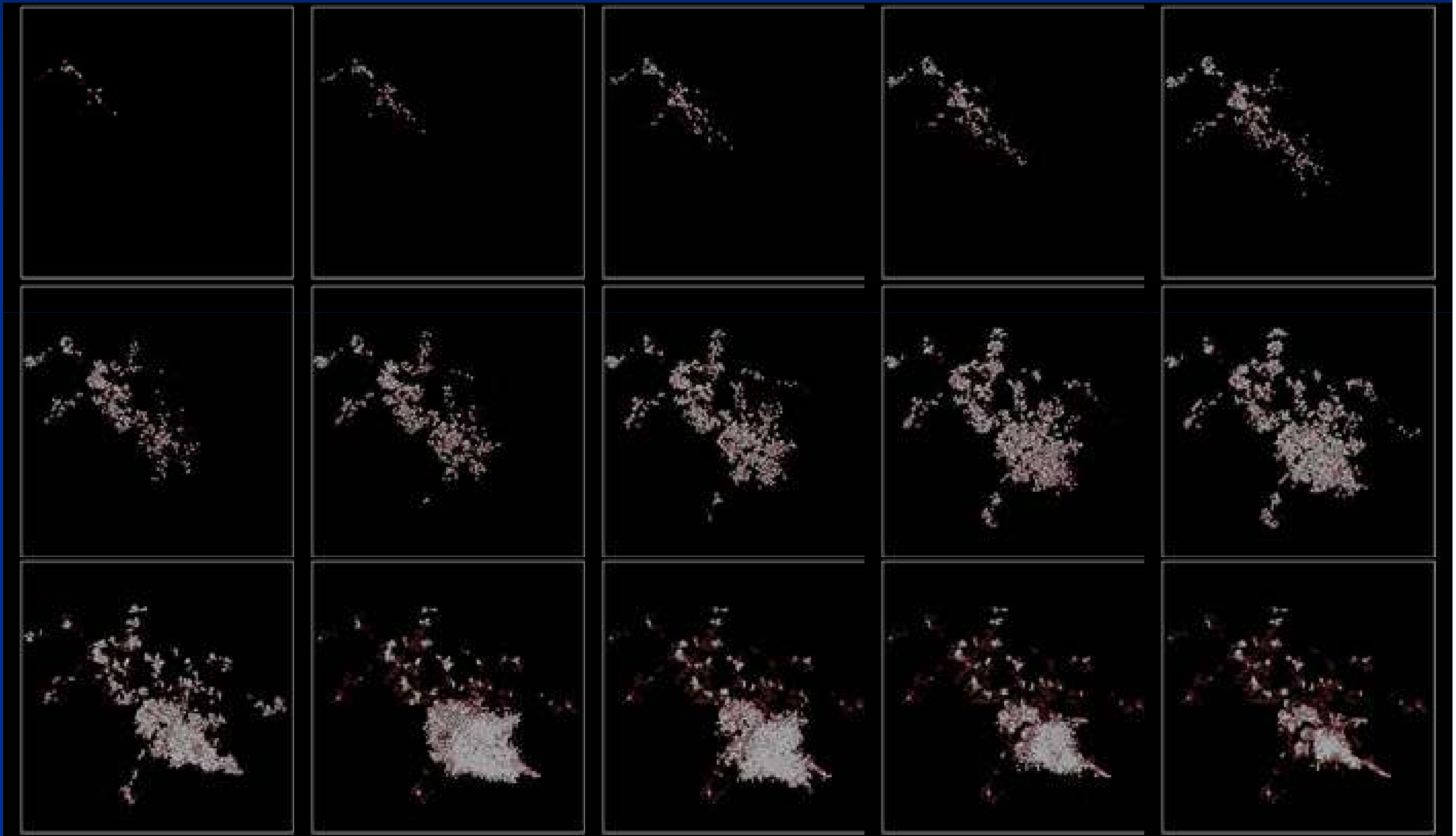
**What is the nature of the process of resistance to amorphization by radiation damage?**

# Modelling resistance to amorphization by radiation damage



- 100 keV in rutile $TiO_2$, ~5-10 mln atoms, MD box size is ~500 Å
- 512-1024 HPCx parallel processors

# Look at the process in detail
## Rutile $TiO_2$

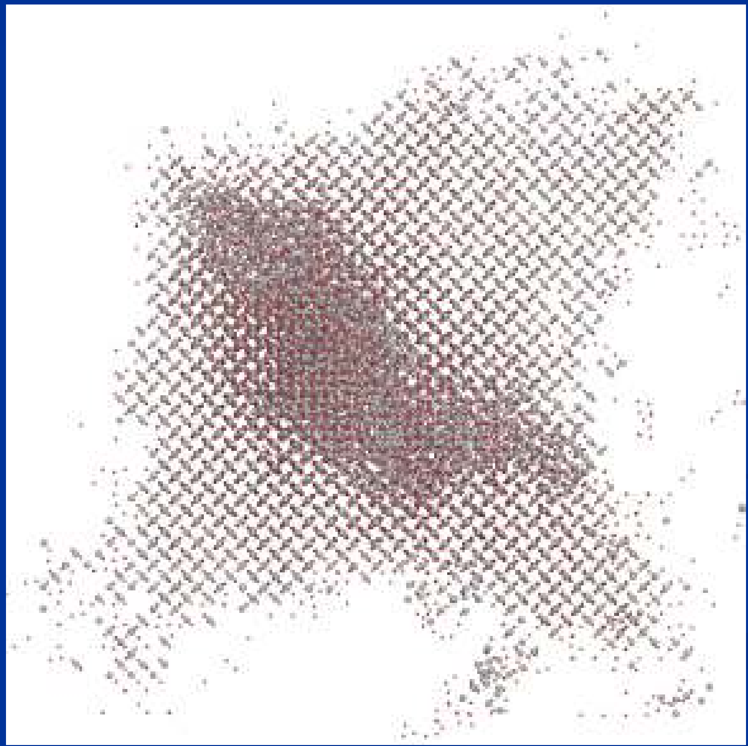


**Two types of relaxation:**

1. Elastic relaxation. Reversible.

2. Relaxation and recovery of the true structural damage

Both happen on the few picosecond timescale.



# Resistance to amorphization

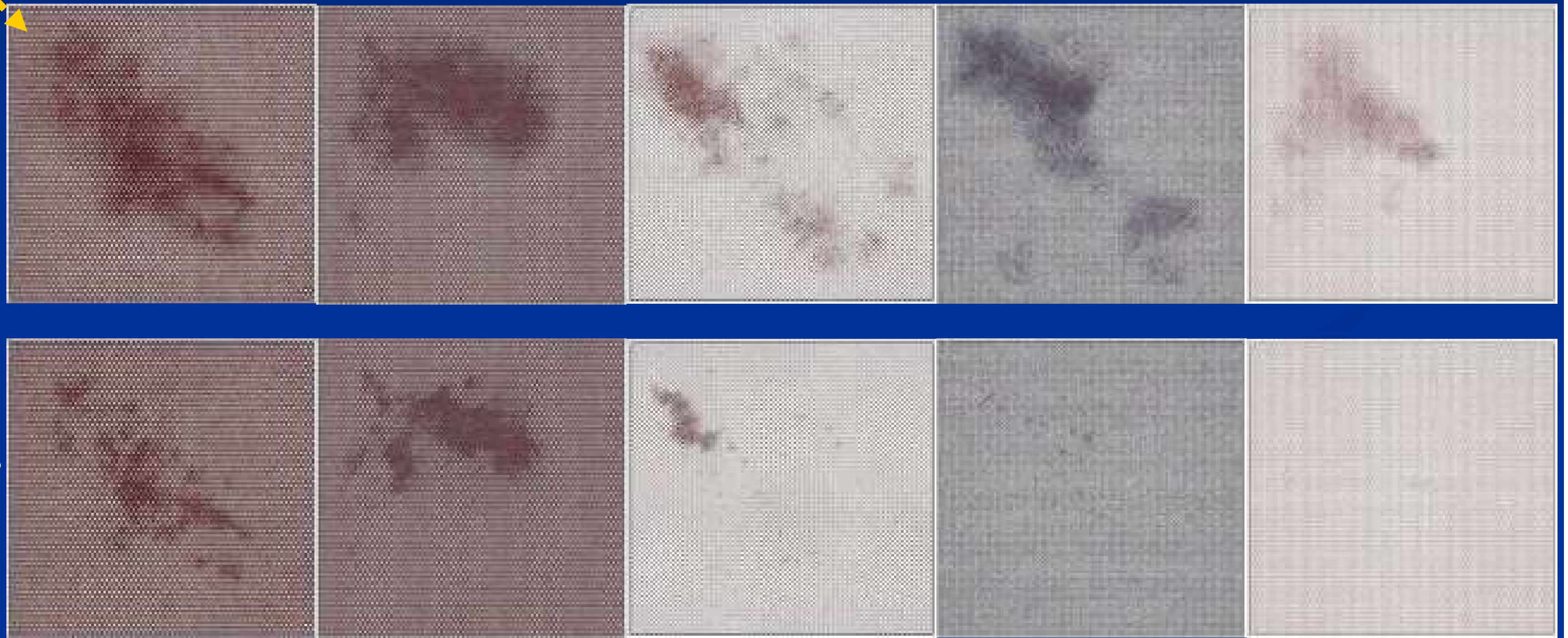
50 keV U recoil

Poor recovery | Intermediate recovery | Good and perfect recovery

$SiO_2$ | $GeO_2$ | $TiO_2$ | MgO | $Al_2O_3$

t=1 ps

t=5, 50 ps



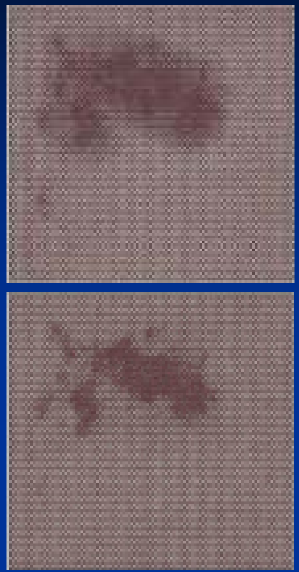
K Trachenko et al, Phys. Rev. B, 2006.

MD simulations **reproduce** experimental behaviour of resistance to amorphization.
(activation barriers correlate with the curvature of the potential at equilibrium)

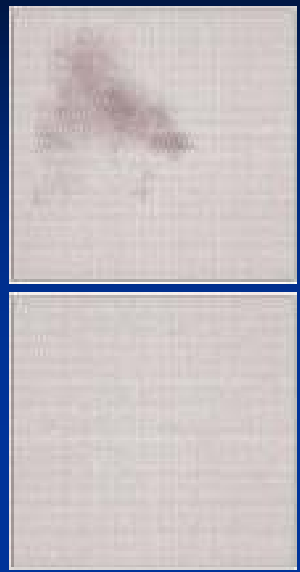
# Resistance to amorphization

t=1 ps

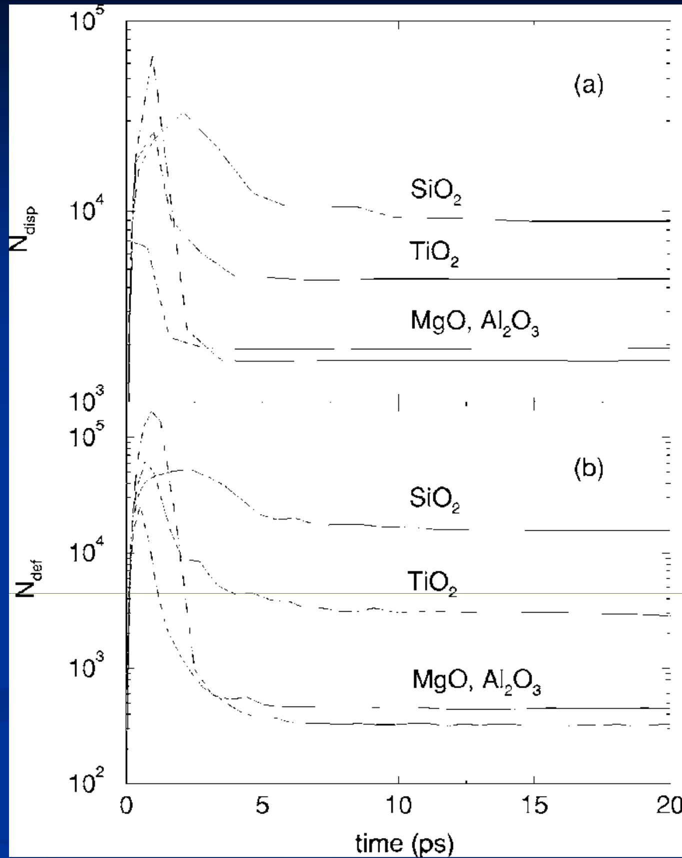
t=5, 50 ps



$GeO_2$



$Al_2O_3$




- Time scales of damage recovery: several ps

- Correlate the details of interatomic potentials with damage recovery: damage increases with the **stiffness** of O-O interaction

## Resistance to amorphization

MD simulations **reproduce** experimental behaviour of resistance to amorphization.

MD simulations can be used to **predict highly resistant materials** (e.g. $ZrO_2$, $Gd_2Zr_2O_7$) where resistance to amorphization operates on the time scale of picoseconds

## Current work and future plans: fusion!

Simulate radiation damage of energies relevant to fusion reactors: high radiation fluxes (200 dpa and 500 keV-1 Mev Fe recoil atoms from 14 MeV neutrons)

- No experiment possible other than fusion reactor. MD simulations are therefore important.
- These energies were not studied before, yet are important to simulate
- Need **up to 1 billion atoms** in a MD box. Doable with 16,382 HECToR processors. The size of this system is about **0.2 micrometers** ! ($2^{nd}$ in the world after Lawrence Livermore people)
- Study the effects of temperature, pressure, important mechanical properties, deformation, elasticity etc
- Possibly exciting new effects, as new energy and length scales (μm) are approached

## Current and future work on fusion

Simulate radiation damage of energies relevant to fusion reactors:
500 keV – 1 Mev Fe recoil atoms from 14 MeV neutrons

- One configuration of 250 mln atoms is about 100 Gb, 1000-frame history file is 100 Tb !
  We will be facing and solving new interesting challenges:
  writing speed and storage (parallel writing, analyze on the fly what we can)
- Common to all future MD simulations of very large sizes. The appetite for large systems approaching μm is growing: shock, fracture, initiation of micro-cracks, micro-structural changes and interfacial effects, macromolecules, biological systems of wide ranges

# Current and future work on fusion

Simulate radiation damage of energies relevant to fusion reactors:
500 keV – 1 Mev Fe recoil atoms from 14 MeV neutrons

- Include electronic energy loss in the simulation - in collaboration with Ilian Todorov (Daresbury) and Dorothy Duffy (UCL).
- Exciting new effects, as new energy and length scales (μm) are approached

Ongoing work, supported by EPSRC grants:

1. "Development of high-performance software"
2. Impact QM: collaboration with the UK National Nuclear Laboratory
3. "Pathways to Impact": collaboration with the Culham Centre for Fusion Energy

First attempt:

~ 100 million -1 billion atoms

~2048-8192 parallel HECToR processors

250 keV Fe recoil in iron

System size ~ 1000 Angstrom

Thank you