SAND2011-7568 C

SAND2011-7568C

TEM with in situ Ion Irradiation: Historical Perspective

J.A. Hinks

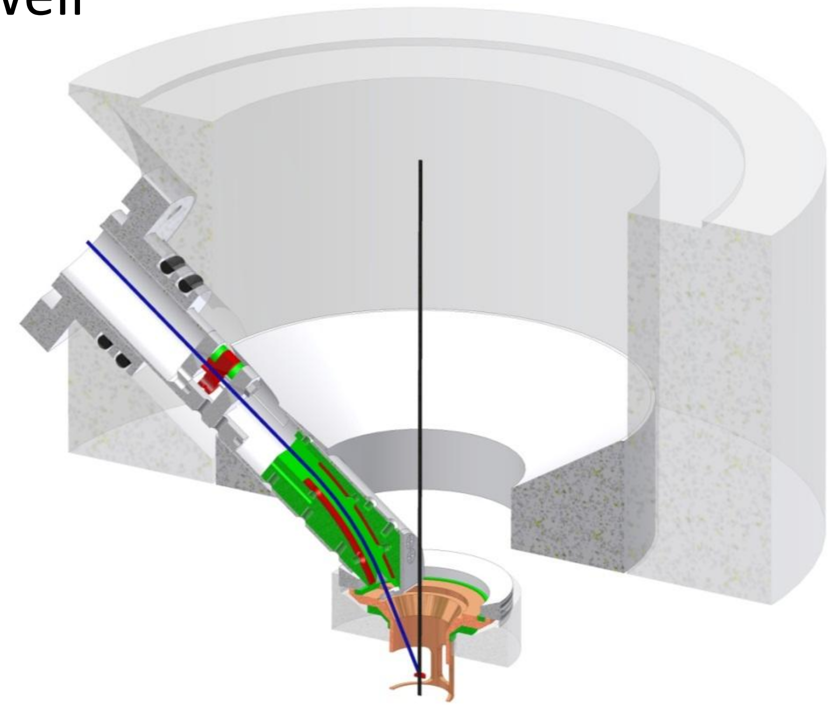
Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

University of Huddersfield

Second Workshop on the Use of In Situ TEM / Ion Accelerator Techniques
Albuquerque, New Mexico, USA, 6th June 2011

Talk Outline

- How I ended up researching this history
- TEM with in situ ion irradiation – stuff you already know!
- The very first experiments
- First integrated systems at Harwell
- Through the decades
- Some facilities
- Choosing equipment
- Bring it together: interfacing
- The future
- Our laboratory move!



Second Workshop on the Use of In Situ TEM / Ion Accelerator Techniques
Albuquerque, New Mexico, USA, 6th June 2011

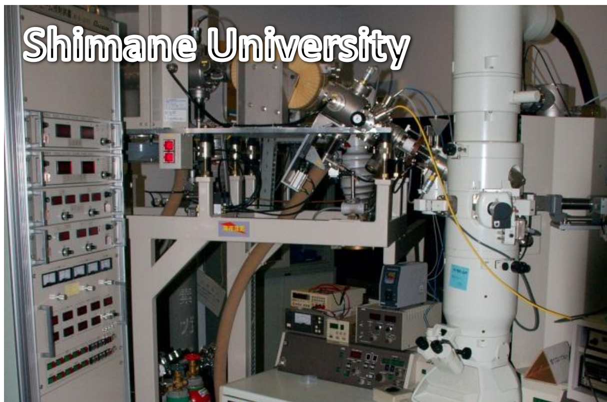
How I Ended up Researching this History

- In 2008 Steve Donnelly and I organised the first workshop
- The idea was to bring together all the early-stage researchers in the field to form at an early stage a network of experience that most researchers come to enjoy later in their careers

Second Workshop on the Use of In Situ TEM / Ion Accelerator Techniques
Albuquerque, New Mexico, USA, 6th June 2011

How I Ended up Researching this History

- Visited twelve institutions including six facilities
- Sought out early-stage researchers and toured facilities
- Gathered lots of information and photographs!



How I Ended up Researching this History

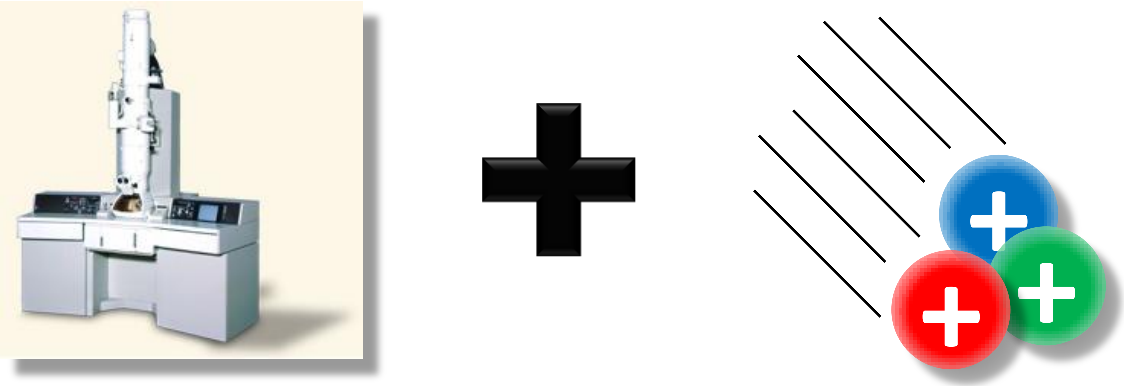
- Whilst seeking out existing facilities gather papers on lots of decommissioned ones – so decided to put it all into a review:

J.A. Hinks *NIMB* **267** (2009) p3652

- But I was not the first to do this something like this:

S. Ishino *J. Nucl. Mater.* **206** (1993) p139
S. Ishino *J. Nucl. Mater.* **251** (1997) p225
C.W. Allen et al *Trans. Mater. Res. Soc. Jpn.* **17** (1994) p93
C.W. Allen *Ultramicroscopy* **56** (1994) p200
C.W. Allen et al *Microsc. Res. Tech.* **42** (1998) p255
R.C. Birtcher et al *J. Mater. Res.* **20** (2005) p1654
Y. Serruys et al *C.R. Phys.* **9** (2008) p437

Preaching to the Converted: Why TEM with in situ Irradiation?

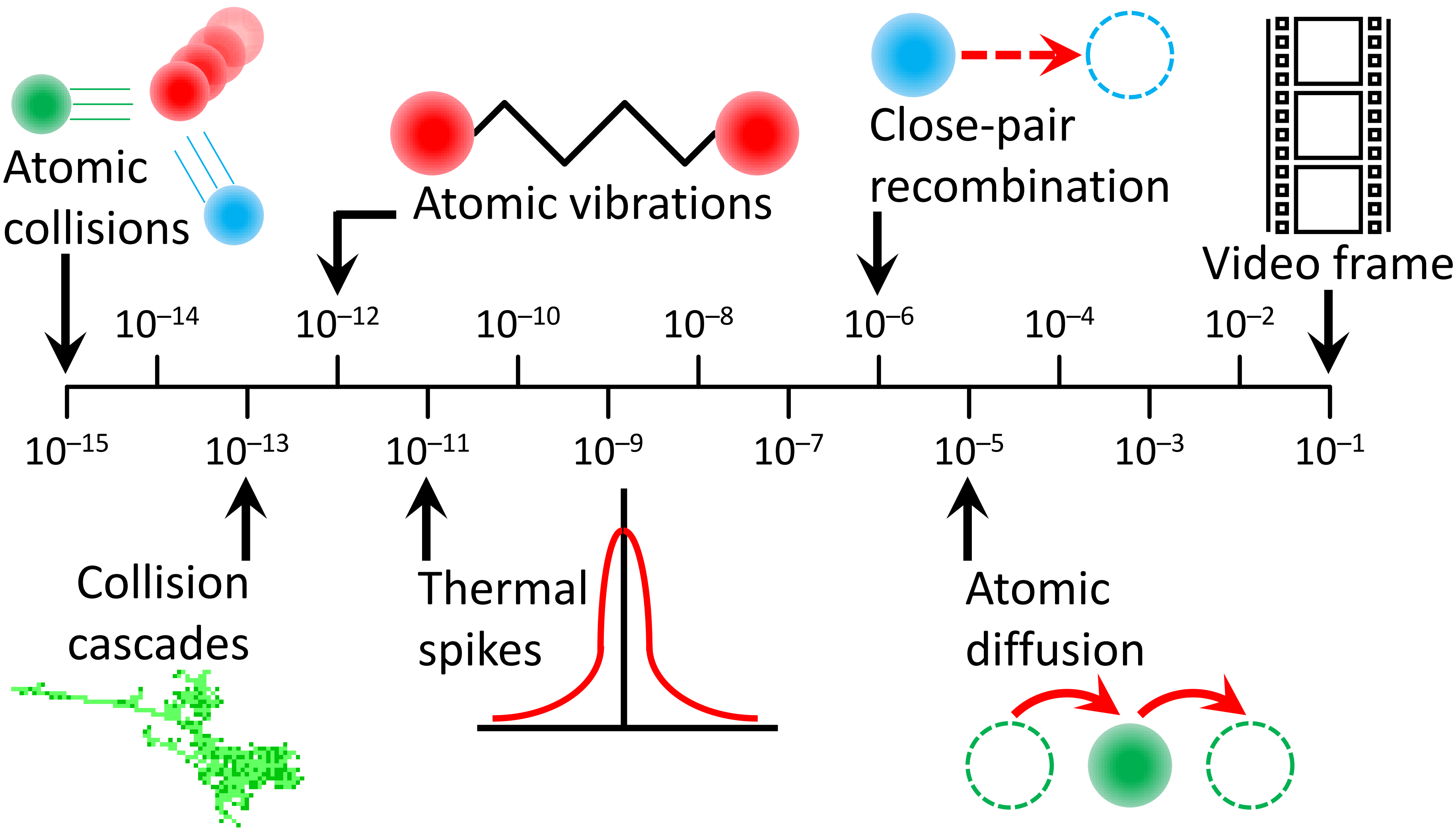


- TEM: nanoscale exploration of internal structure of materials
- The effects of irradiation are dynamic – competing processes
- Ex situ studies give access only to the end-states
- Can observe effects as they occur within the sample
- Can vary and maintain parameters such as temperature during both irradiation and measurement

Preaching to the Converted: Advantages over ex situ Studies

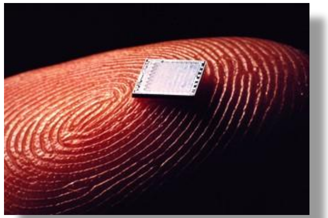
- No need to remove sample from TEM for each irradiation step – can gather data much more quickly and avoid contamination
- Can also record data continuously as irradiation is performed
- Much easier to maintain temperature during both irradiation and observations
- Maintain sample area and observe sudden/significant changes
- Can maintain identical TEM imaging conditions

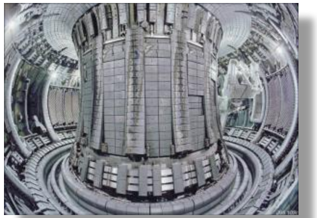
Preaching to the Converted: Timescales

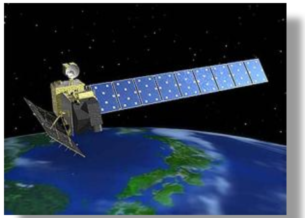


Second Workshop on the Use of In Situ TEM / Ion Accelerator Techniques
Albuquerque, New Mexico, USA, 6th June 2011

Preaching to the Converted: Applications







- Investigations into any materials subjected to radiation damage
- Materials for nuclear fission (current and GenIV), nuclear fusion (especially for plasma facing components) and ceramics for nuclear waste storage
- Semiconductor processing and damage to devices used in irradiating environments
- Nanotechnology – e.g. effects of ion irradiation on nanowires
- Fundamental research into underlying physics and atomistics

The Very First Experiments

Ion Damage to Metal Films inside an Electron Microscope

By D. W. Pashley and A. E. B. Presland†
T.I. Research Laboratories, Hinxton Hall, Cambridge

[Received January 30, 1961]

Abstract

The origin of small spot and loop features observed in electron micrographs of thin evaporated single-crystal gold films has been investigated. It is found that the features are introduced into the specimens whilst they are being examined in the electron microscope. Conclusive evidence is given to show that they arise from bombardment by energetic negative ions emitted from the tungsten filament of the electron gun, although the ions have not been identified. The rate of ion damage can be considerably increased by coating the tungsten filament with a standard oxide emitter.

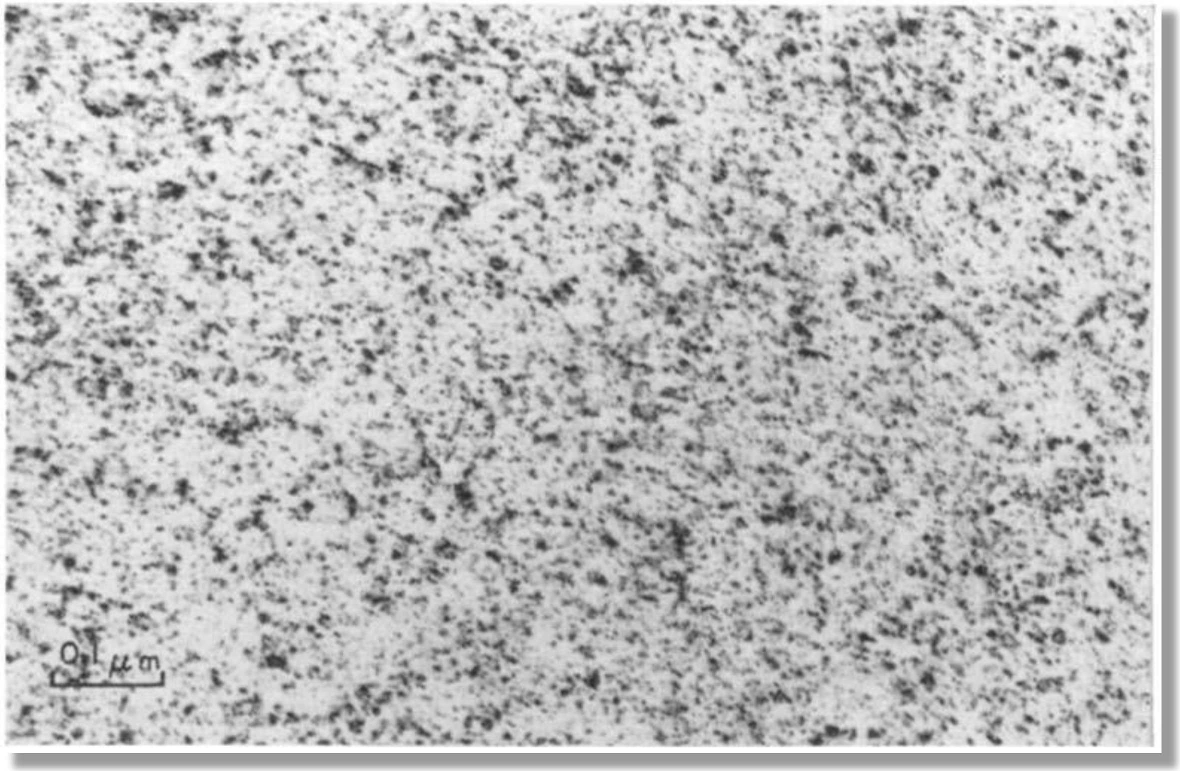
The annealing characteristics of the damage have been studied. Above about 300–350°c the majority of the spot features anneal out, and ion damage above about 350°c consists of the formation of small tetrahedra of stacking fault.

The ion damage is assumed to be very similar in character to that produced by primary knock-ons resulting from irradiation by other particles (e.g. neutrons).

Philosophical Magazine **6 (68)** (1961) p1003

The Very First Experiments

- Black spot damage was observed to form in Au samples in a Siemens Elmiskop I 100kV TEM at Tube Investment Research Laboratories, UK



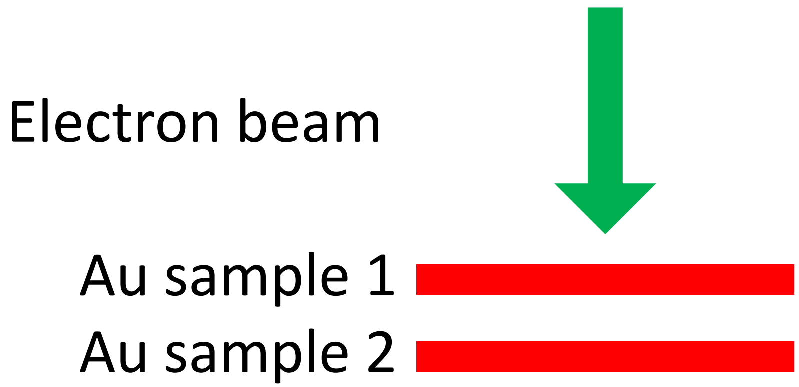
L.M. Howe et al *Acta Metallurgica* **14** (1966) p 801

The Very First Experiments

- To rule out agglomeration of pre-existing point defects under electron beam samples were annealed ex situ – still produced damage
- From RT to 300°C got black spot damage and 350 to 400°C got tetrahedral stacking faults
- Suspected negative ion emission from tungsten filament of electron gun – most probably O^-
- Used deflection coils above sample position to deflect electrons but not ions (if present) – still produced damage

The Very First Experiments

- As 100 keV ions of probable species would nearly all be stopped in the Au sample thickness, they did a further test:



- By changing the strength of the objective lens they could focus on either one sample or the other – they discovered they got damage in the first sample but not the second

- Concluded that they were indeed performing TEM with in situ ion irradiation! Also, first record of in situ screening effects!

The Very First Experiments

"The direct observation of ion damage in the electron microscope thus represents a powerful means of studying radiation damage"

D.W. Pashley and A.E.B Presland *Philosophical Magazine* **6 (68)** (1961) p1003

The Very First Experiments

- Our story now moves to the Atomic Energy of Canada Limited, Chalk River, Canada
- Following on from the work of Pashley, Presland and Menter, experiments were done by Howe, Gilbert, Piercy and Parsons
- They also used a Siemens Elmiskop I but with deliberately coated filaments to create higher ion fluxes

Howe et al *APL* **3(8)** (1963) p125

Howe et al *APL* **4(6)** (1964) p99

Parsons et al *J. Sci. Instrum.* **(41)** (1964) p773

Howe et al Acta Met. (1966) p801

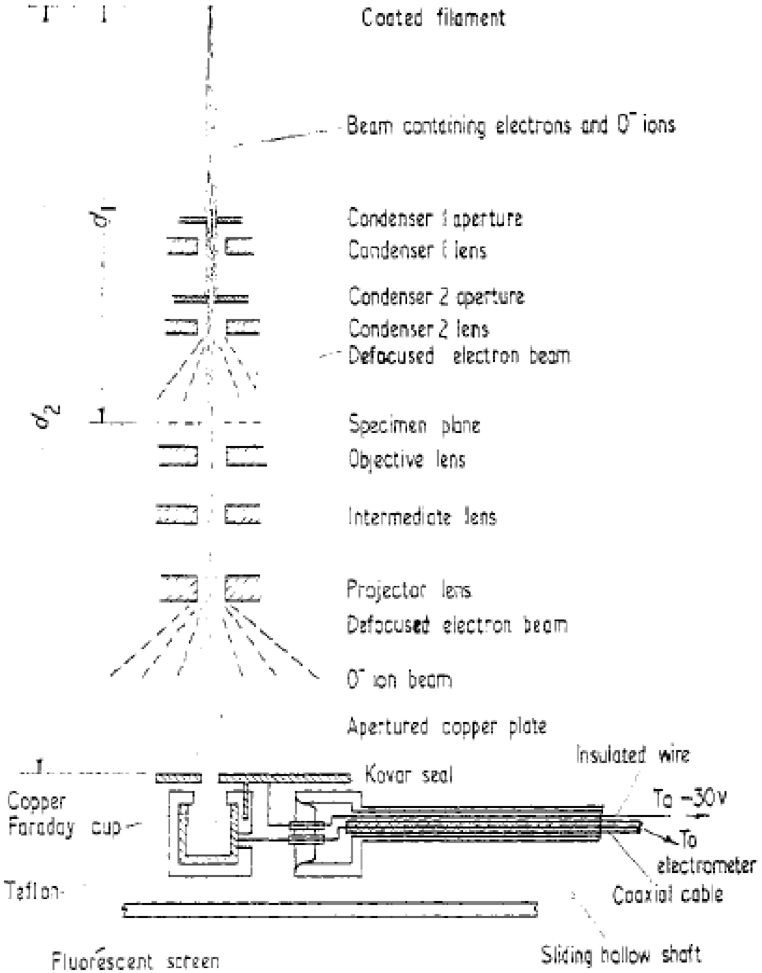
The Very First Experiments

- They had a few other firsts for TEM with in situ ion irradiation

- An adapted cold finger allowed the sample to be cooled to around 30 K

- A Faraday cup was placed just above the viewing screen allowing measurement of the ion flux by configuring the lenses to heavily diverge the electron beam

- *J. Sci. Instrum.* **(41)** (1964) p773



Second Workshop on the Use of In Situ TEM / Ion Accelerator Techniques
Albuquerque, New Mexico, USA, 6th June 2011

First Integrated Systems at Harwell

- The first facility to interface an ion beam with a TEM was reported in 1968 by Thackery, Nelson and Sansom
- It used a JEOL JEM-6A 100 kV TEM and two ion sources
- 30–120 kV heavy ion source
- 10–25 kV glow discharge gun
- Atomic Energy Authority Report 5817 (1968)

United Kingdom Atomic Energy Authority

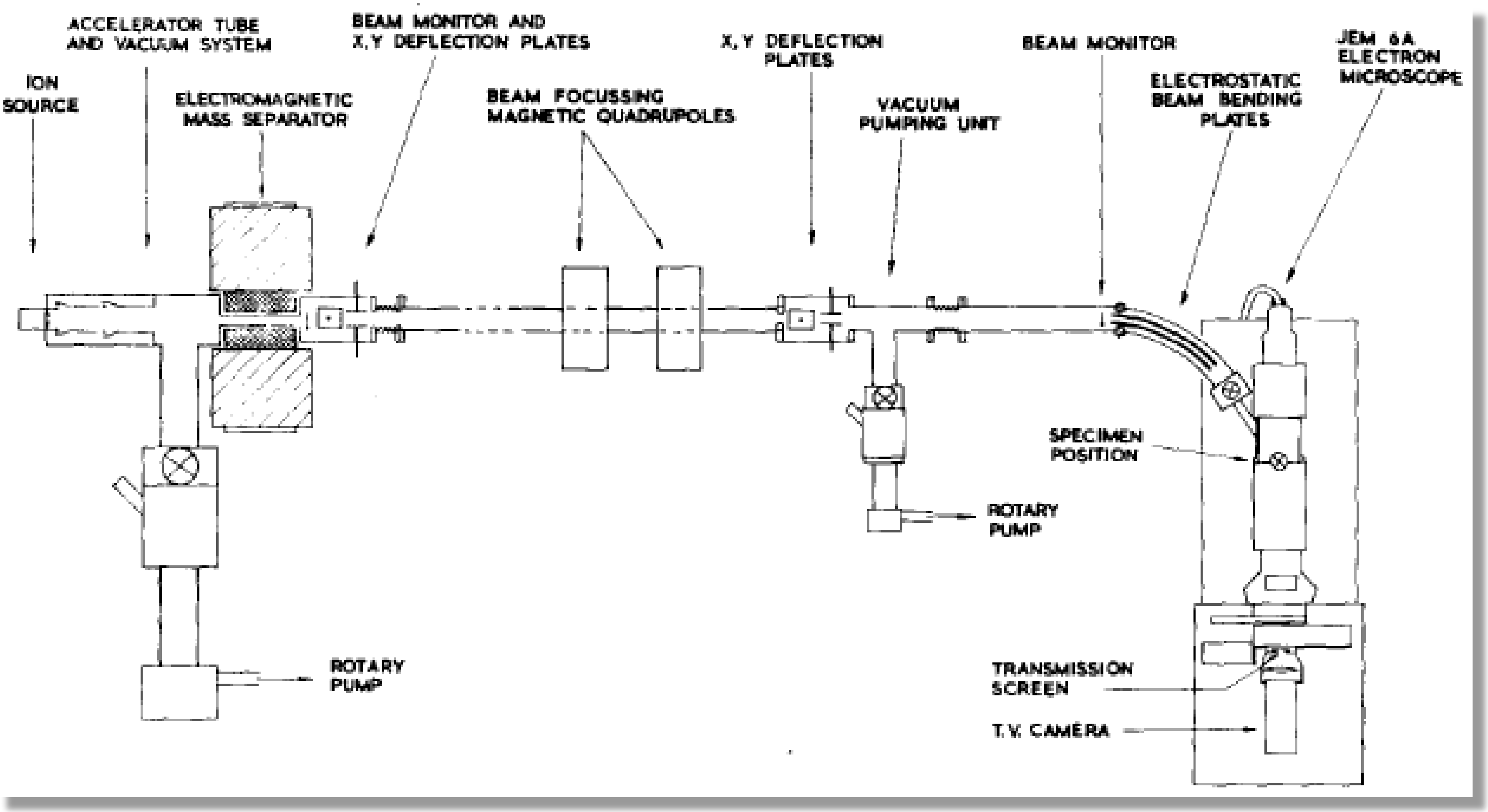
RESEARCH GROUP

Report

A COMBINED HEAVY ION ACCELERATOR-ELECTRON MICROSCOPE FOR THE DIRECT OBSERVATION OF RADIATION EFFECTS

P. A. THACKERY R. S. NELSON H. C. SANSOM

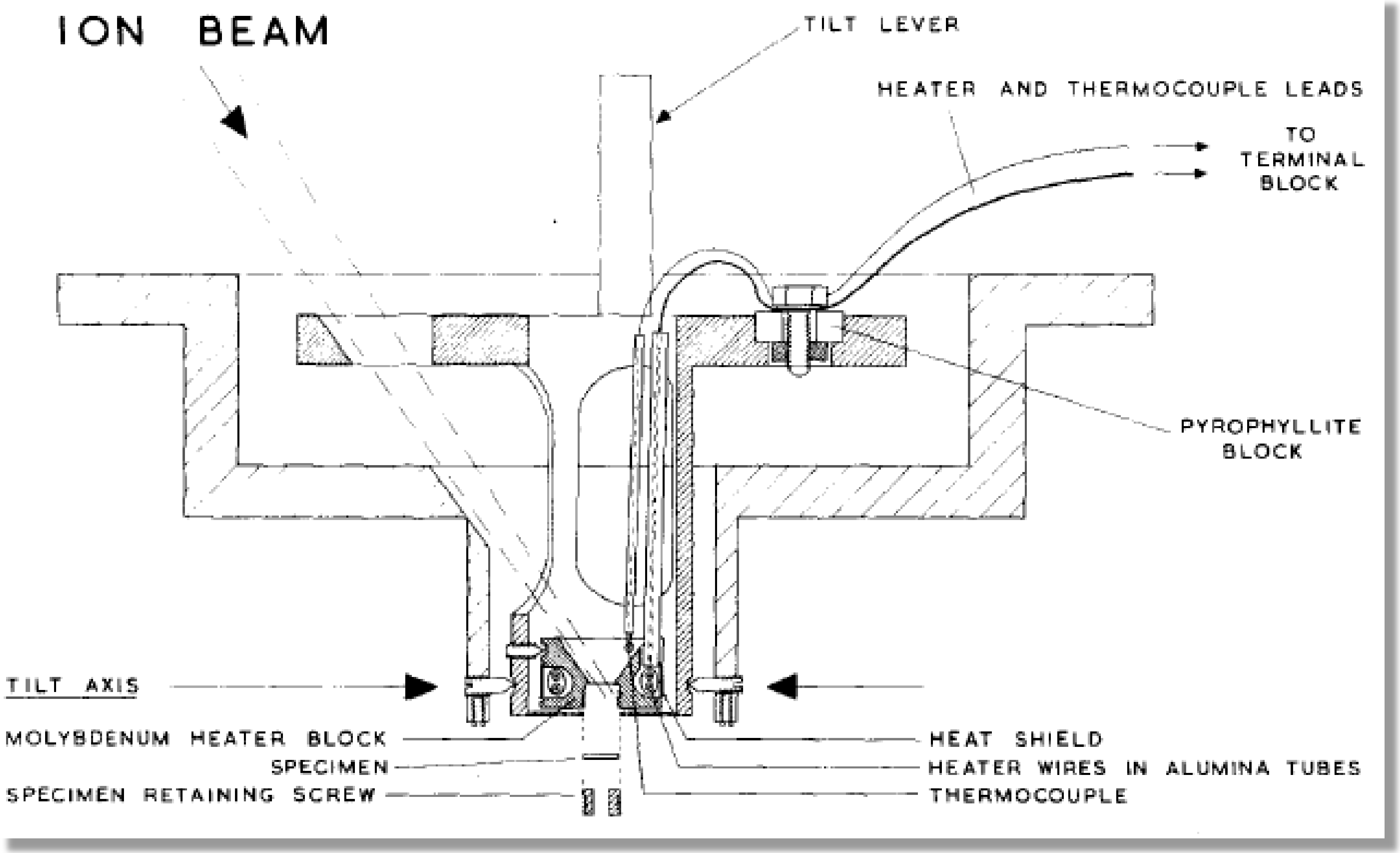
First Integrated Systems at Harwell



Atomic Energy Authority Report 5817 (1968)

Second Workshop on the Use of In Situ TEM / Ion Accelerator Techniques
Albuquerque, New Mexico, USA, 6th June 2011

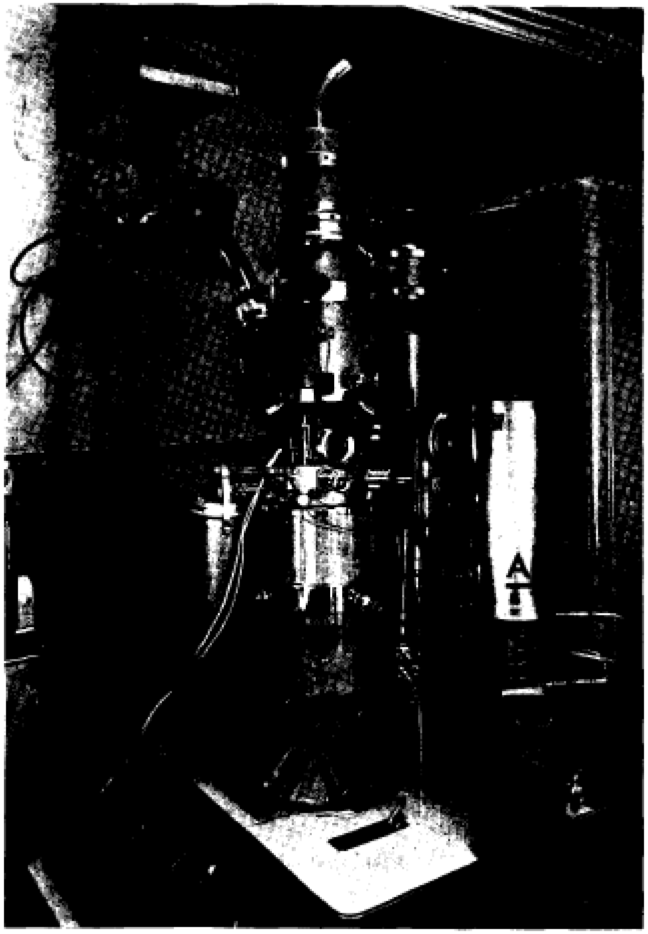
First Integrated Systems at Harwell

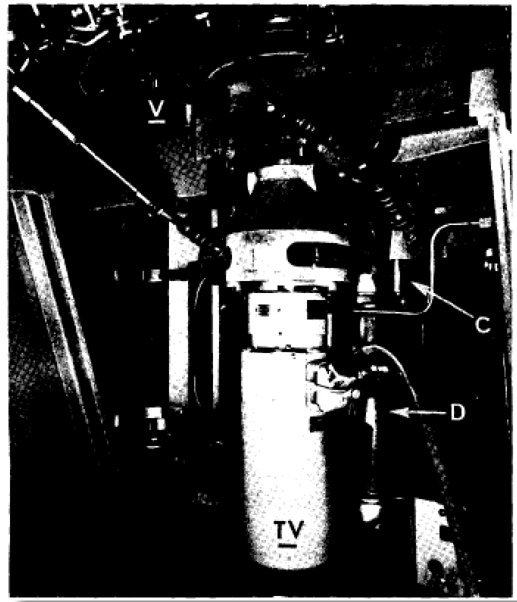


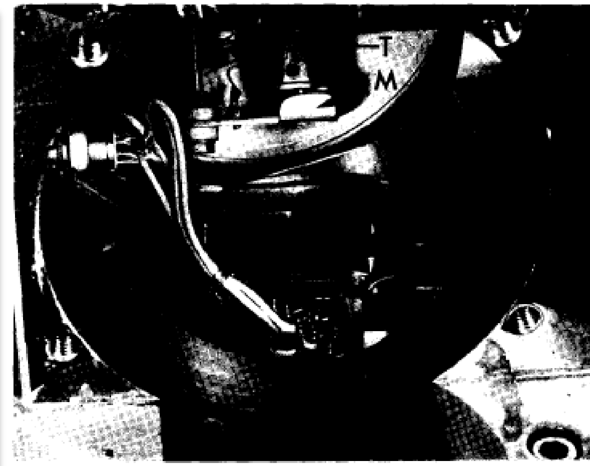
Atomic Energy Authority Report 5817 (1968)

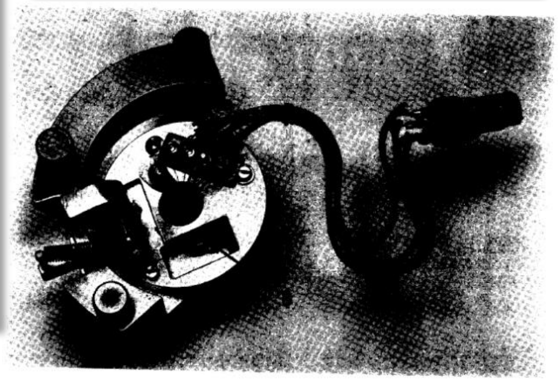
Second Workshop on the Use of In Situ TEM / Ion Accelerator Techniques
Albuquerque, New Mexico, USA, 6th June 2011

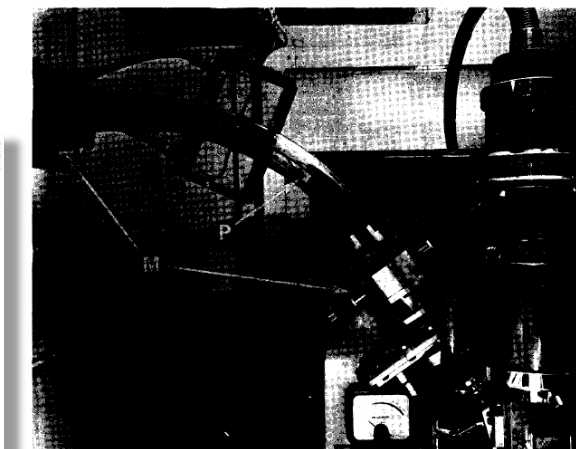
First Integrated Systems at Harwell







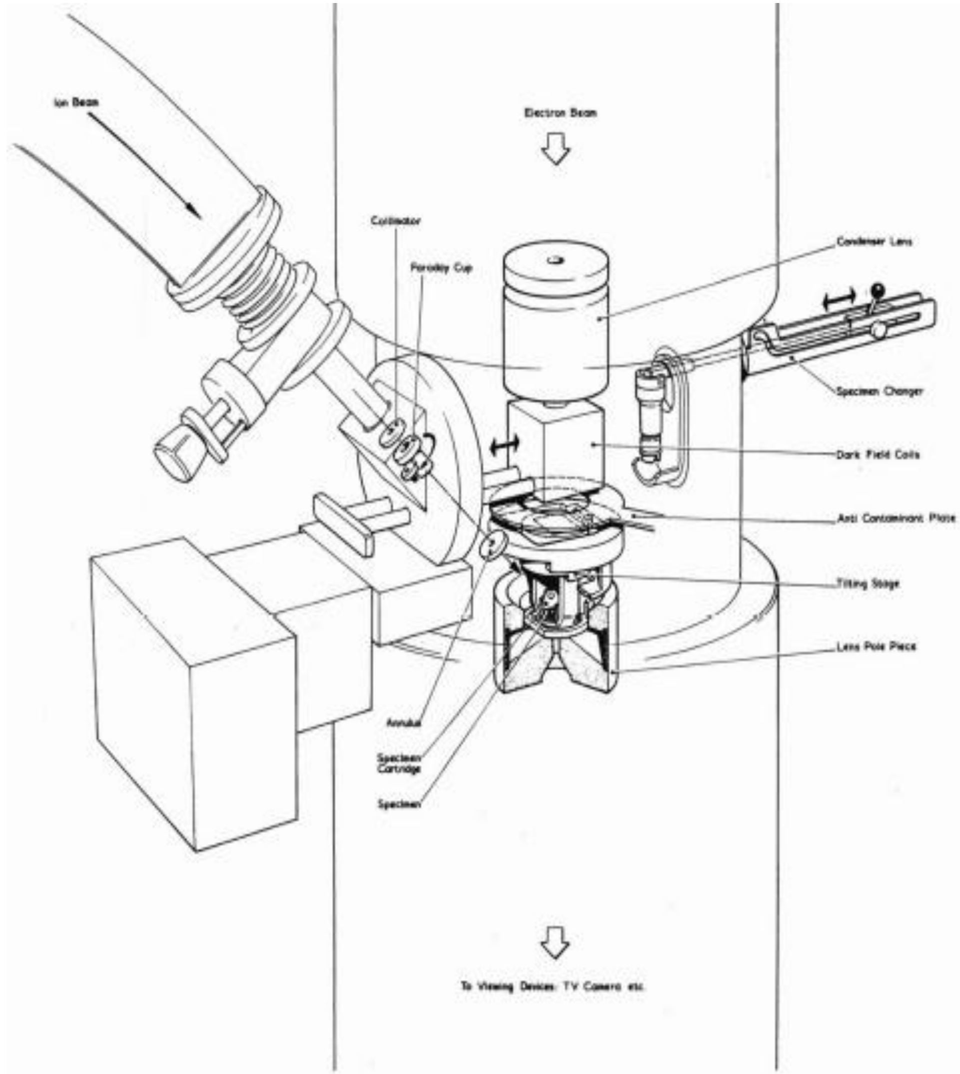




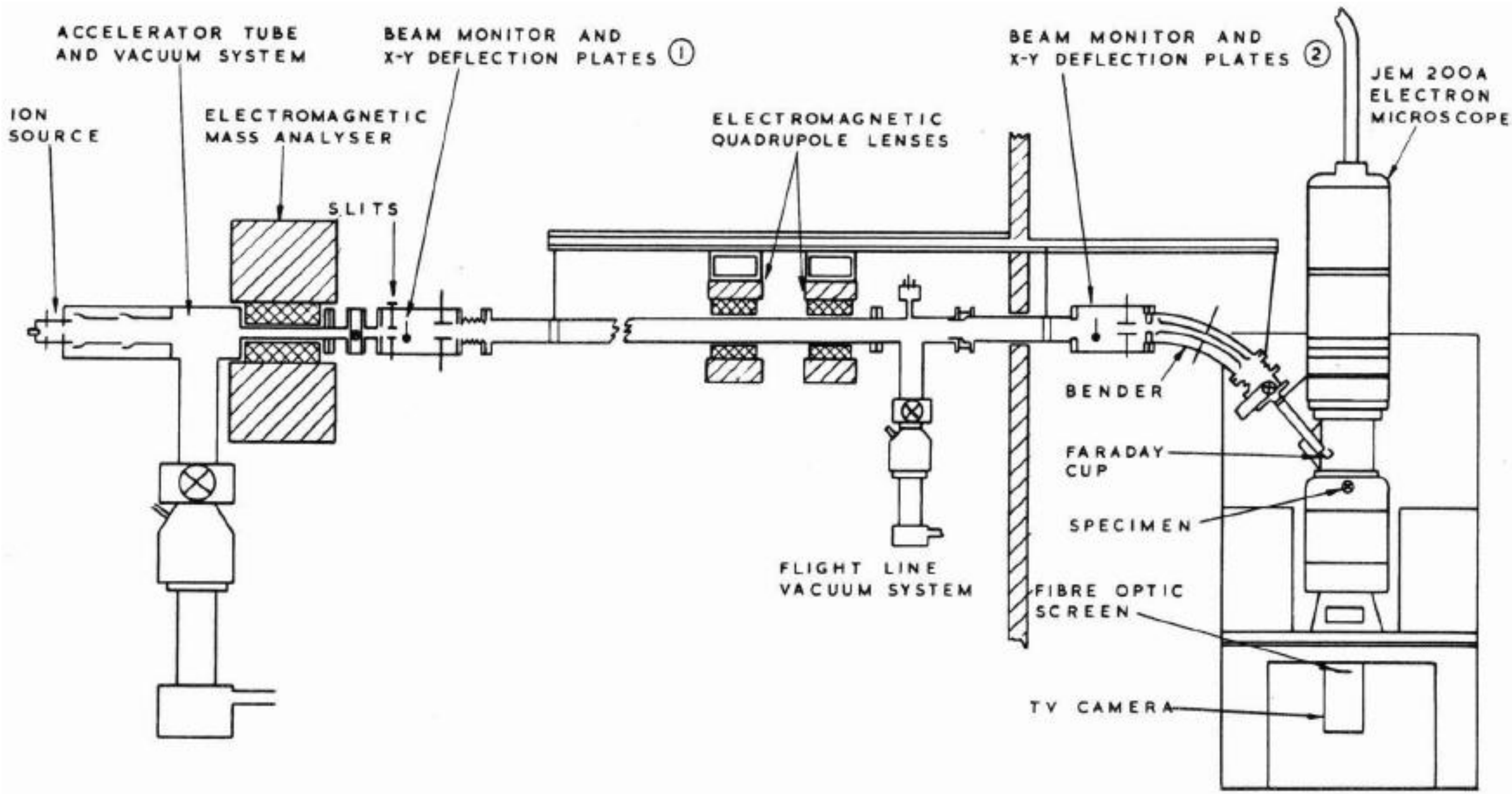
Atomic Energy Authority Report 5817 (1968)

First Integrated Systems at Harwell

- The system was upgraded to a JEOL JEM-200A TEM
- The small ion gun appears to have been removed at this stage
- The beam steering and monitoring were also upgraded
- *Rad. Eff.* **22** (1974) p163



First Integrated Systems at Harwell



Rad. Eff. **22** (1974) p163

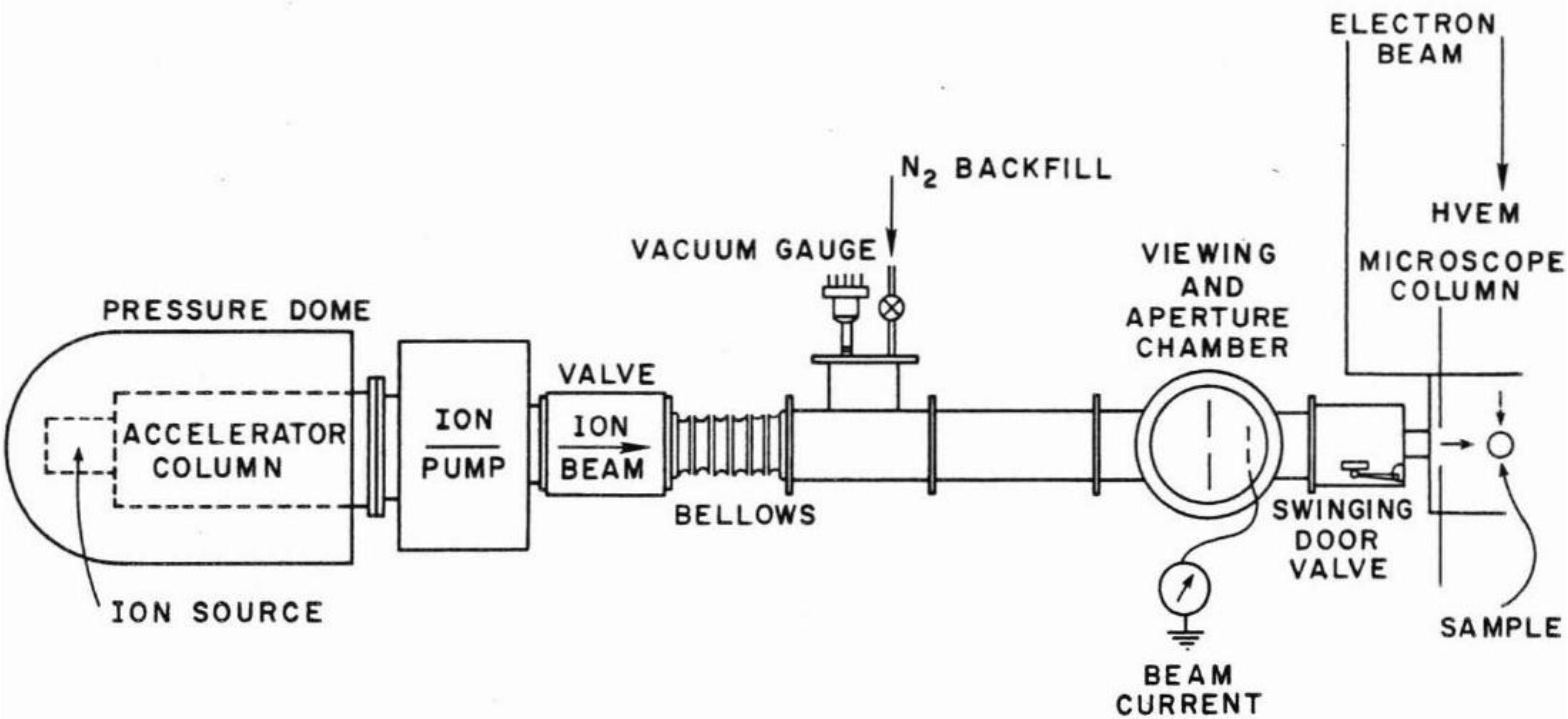
Through the Decades: 1970s

- We are now in the mid 1970s which witnessed the start of a golden age for TEM with in situ ion irradiation with a proliferation of facilities around the world

- Over the next two decades 24 facilities would be built before something of an hiatus in the mid-to-late 1990s

Through the Decades: 1970s

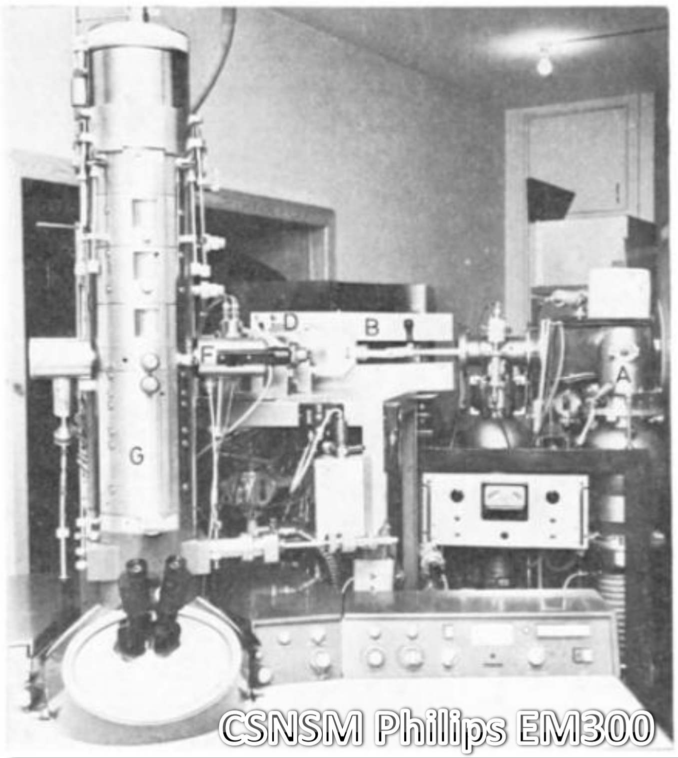
- Leading the way, was the University of Virginia where Jesser built the first facility to be situated outside of the UK

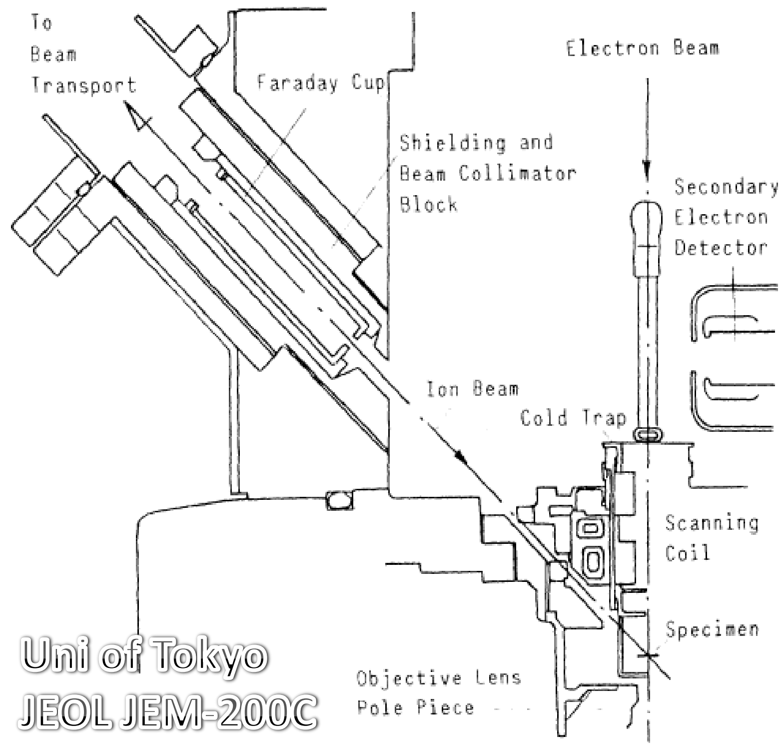


Rad. Eff. **29** (1976) p79

Through the Decades: 1970s

- Too big players, France and Japan, entered the field in 1976 and 1978, respectively





J. Phys. E **11** (1978) p1125 *IEEE Nucl. Sci.* **30(2)** (1983) p1683

Through the Decades: 1980s

- The 1980s started with a second instrument at CSNSM (1980) and the first at Argonne (reported 1981 with TEM installed in 1979)

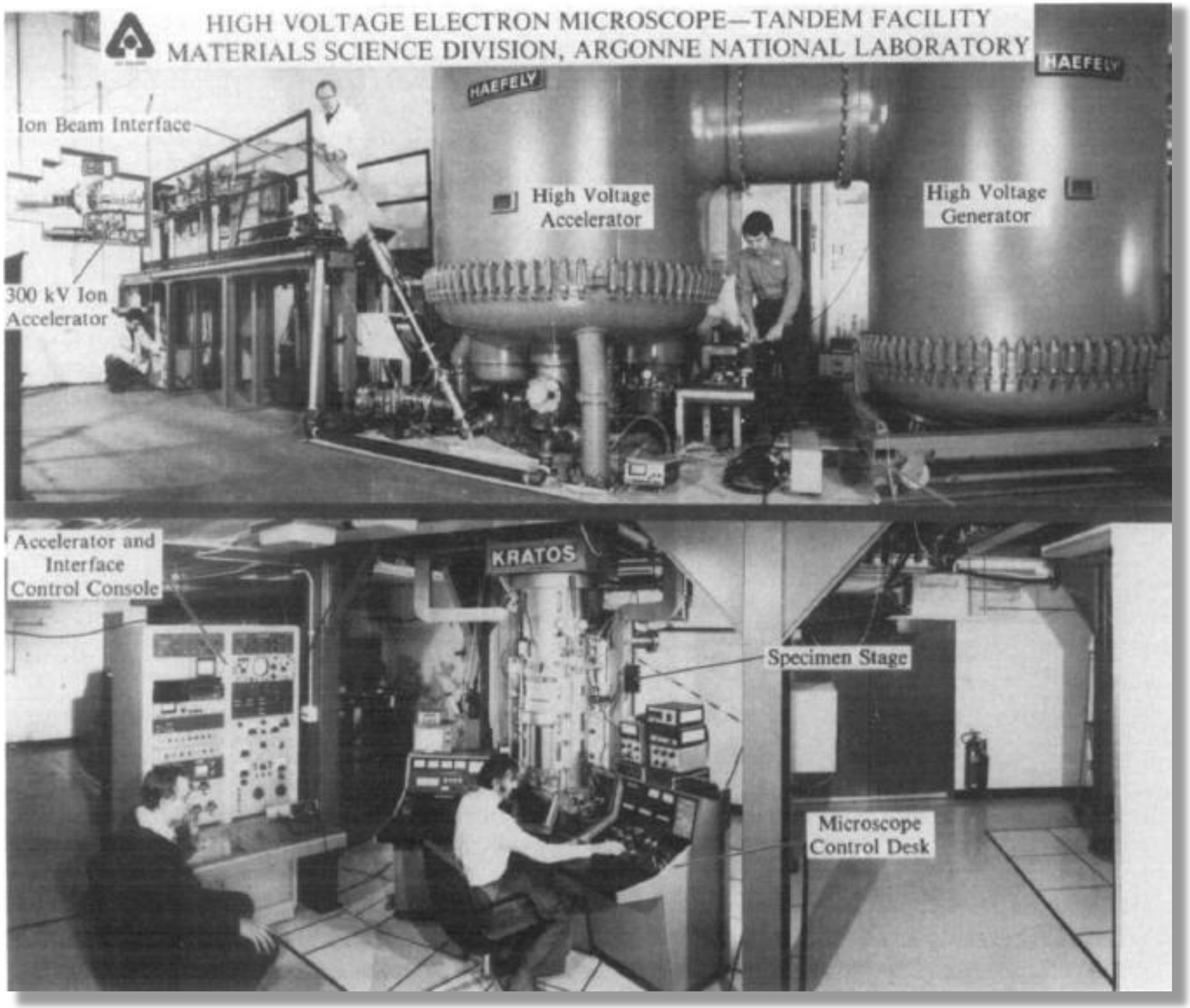
- The Kratos/AEI EM-7 Argonne was the first HVEM to be interfaced with a ion beam

- It could irradiate using either a 300 kV or 2000 MV ion accelerator

- The 300 kV system was upgraded to 650 kV in 1987

- The same beamlines were subsequently used for the IVEM

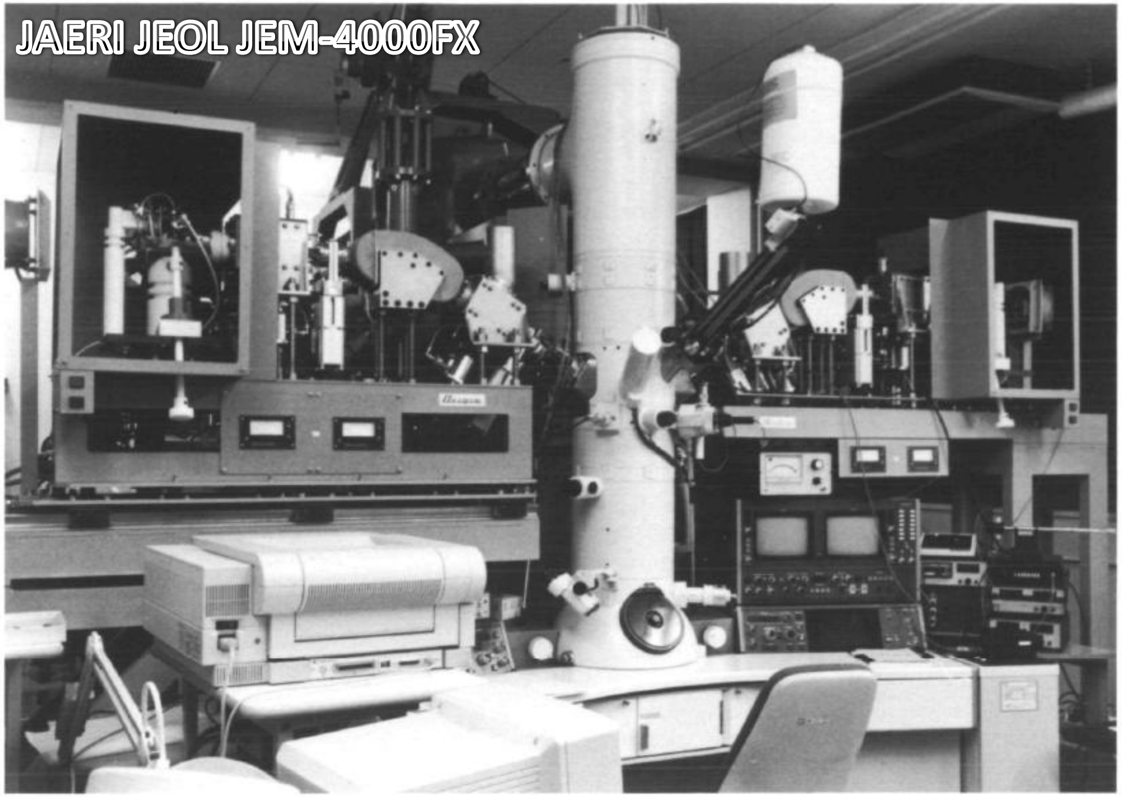
Through the Decades: 1980s



Nucl. Instr. Methods **189** (1981) p211

Through the Decades: 1980s

- However, the rest of decade and early 1990s truly belong to the Japanese who built the next 11 facilities



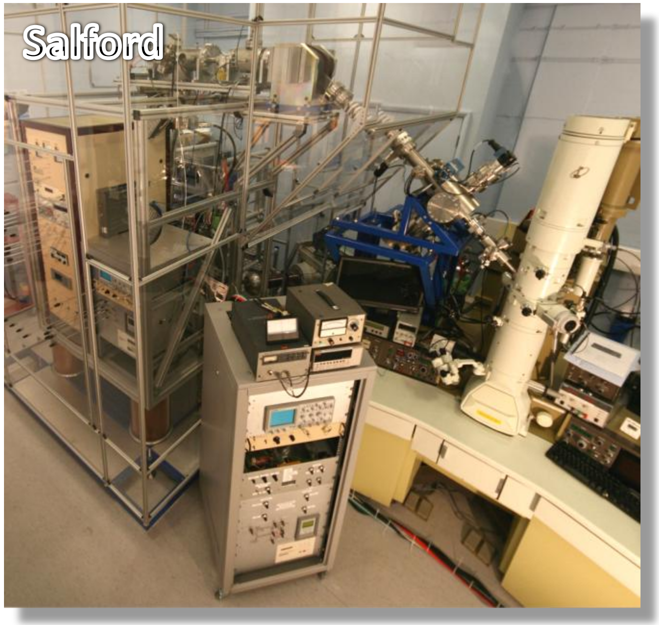
IEEE Nucl. Sci. **30(2)** (1983) p1683

Through the Decades: 1990s

- After the aforementioned Japanese expansion the 1990s continued at a brisk pace
- A further system was built at CSNSM (Orsay) in 1994 and the IVEM was established at Argonne in 1995
- Not to be outdone, the Japanese then built another six facilities from 1995 to 1998

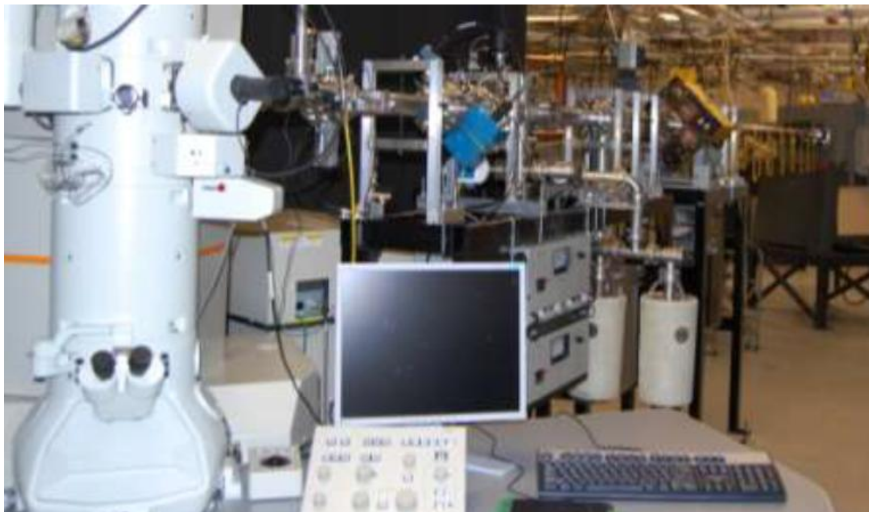
Through the Decades: 2000s

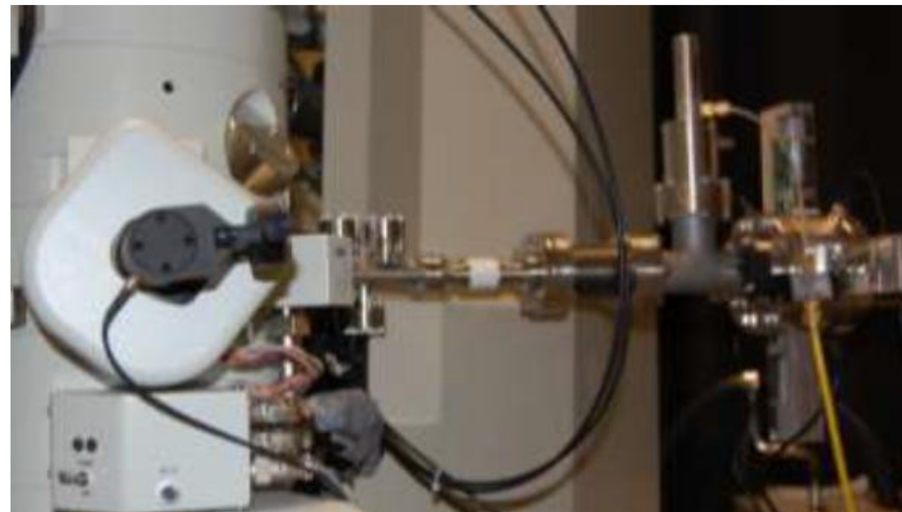
- The next ten years from 1998 to 2008 saw no reports in the literature of new facilities
- However, three were under construction in China, France and the UK



Through the Decades: 2010s

- The latest facility has, of course, been established here at Sandia National Laboratories

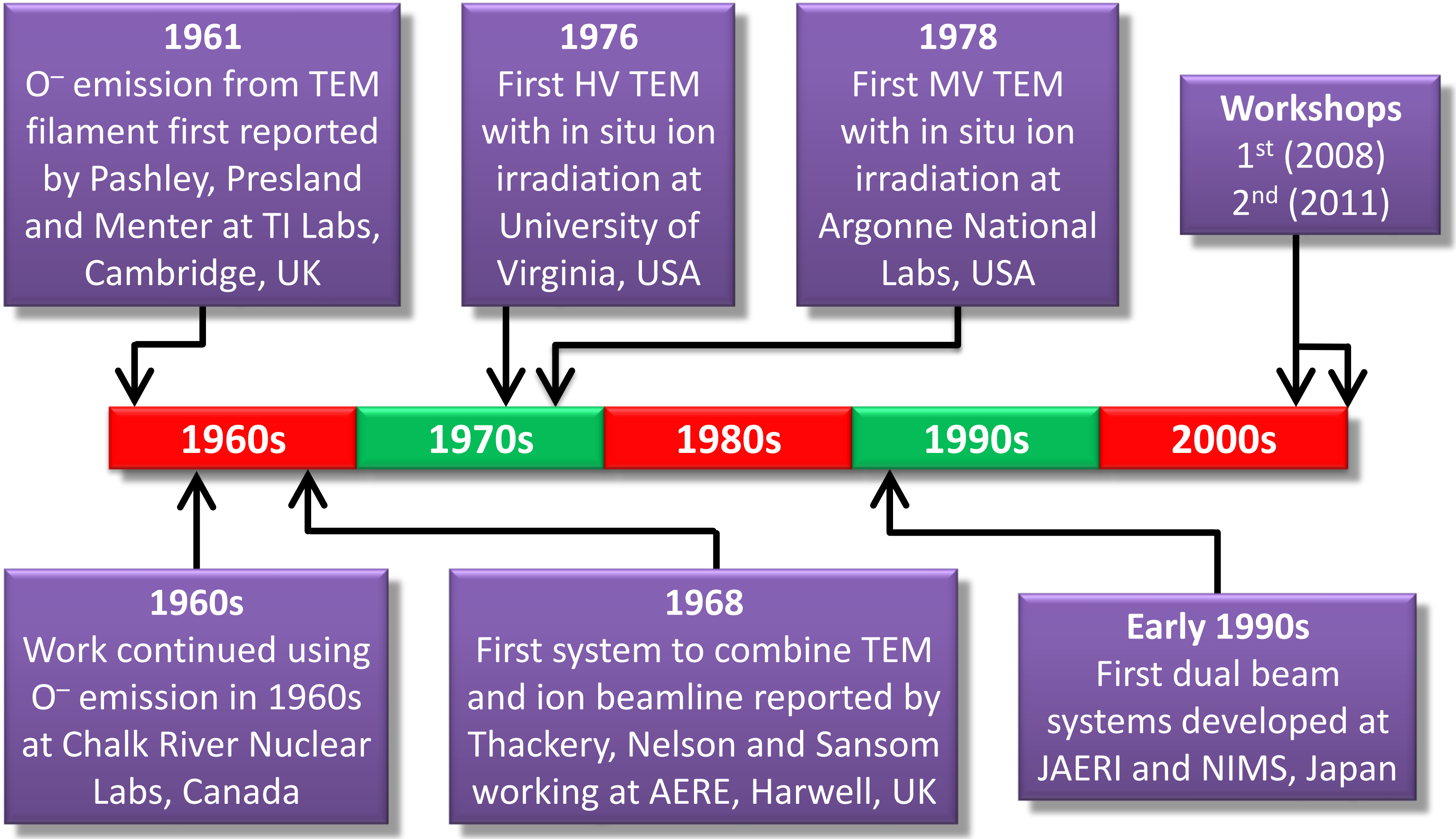




- See: tour of IBL Facilities on Tuesday afternoon and talks in Thursday, Session 9, 0900 to 1030

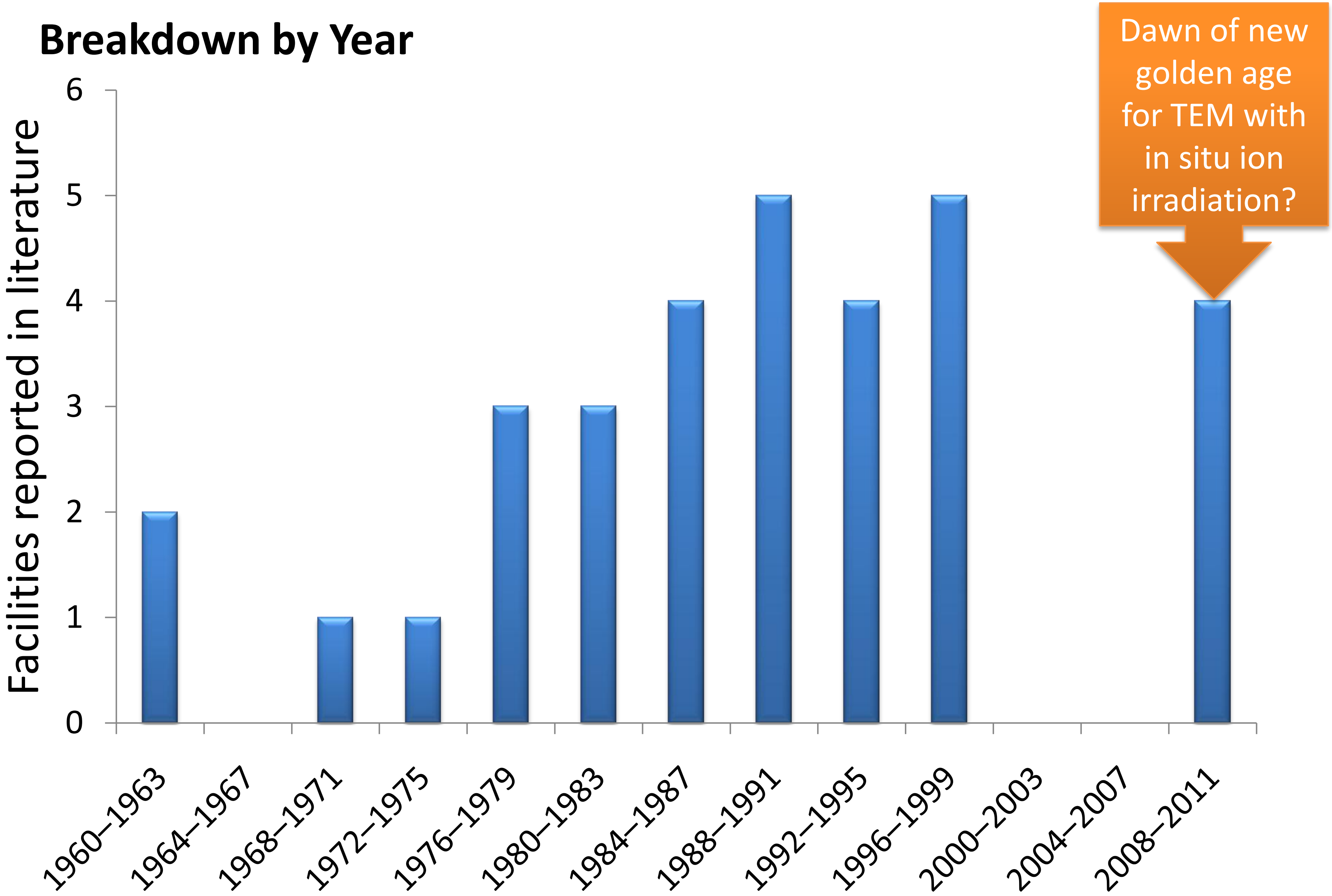
- Images courtesy of Khalid Hattar

Timeline

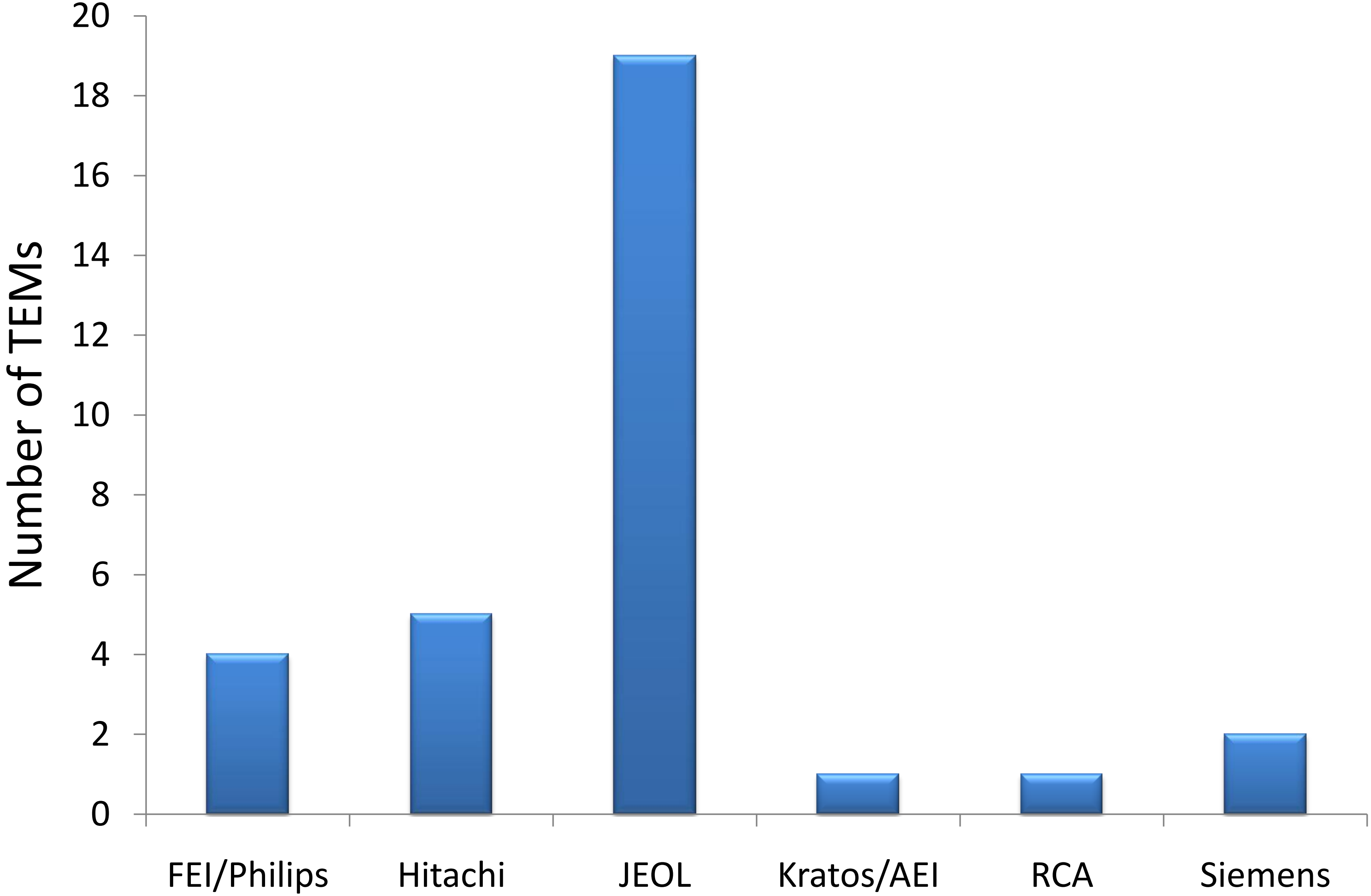


1961
O⁻ emission from TEM filament first reported by Pashley, Presland and Menter at TI Labs, Cambridge, UK
1976
First HV TEM with in situ ion irradiation at University of Virginia, USA
1978
First MV TEM with in situ ion irradiation at Argonne National Labs, USA
Workshops
1st (2008)
2nd (2011)
1960s
1970s
1980s
1990s
2000s
1960s
Work continued using O⁻ emission in 1960s at Chalk River Nuclear Labs, Canada
1968
First system to combine TEM and ion beamline reported by Thackery, Nelson and Sansom working at AERE, Harwell, UK
Early 1990s
First dual beam systems developed at JAERI and NIMS, Japan

Breakdown by Year



Breakdown by TEM Manufacturer



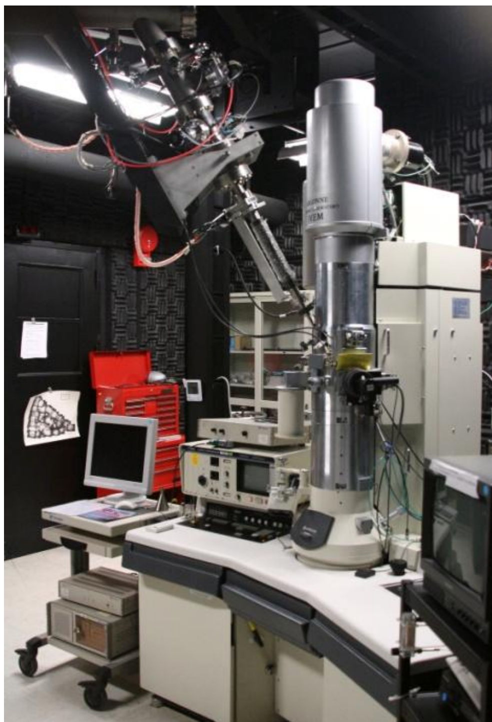
Operational Facilities around the World Today

- Twelve operational facilities at ten different organisations

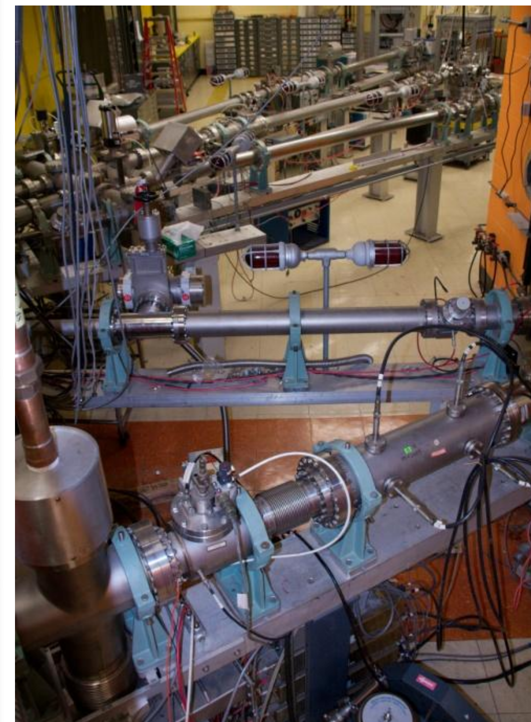
Second Workshop on the Use of In Situ TEM / Ion Accelerator Techniques
Albuquerque, New Mexico, USA, 6th June 2011

Selected Facilities: IVEM at Argonne, Illinois, USA

- Probably the best known facility in the world



Specifications	
TEM	Hitachi H-9000NAR
e-Beam	300 kV
Ion Beam	650 kV or 2 MV
Ion Species	H, He, Ne or Ar Typical flux: 4×10^{10} ions.cm^{-2}.s^{-1}
Angle between Beams	30°

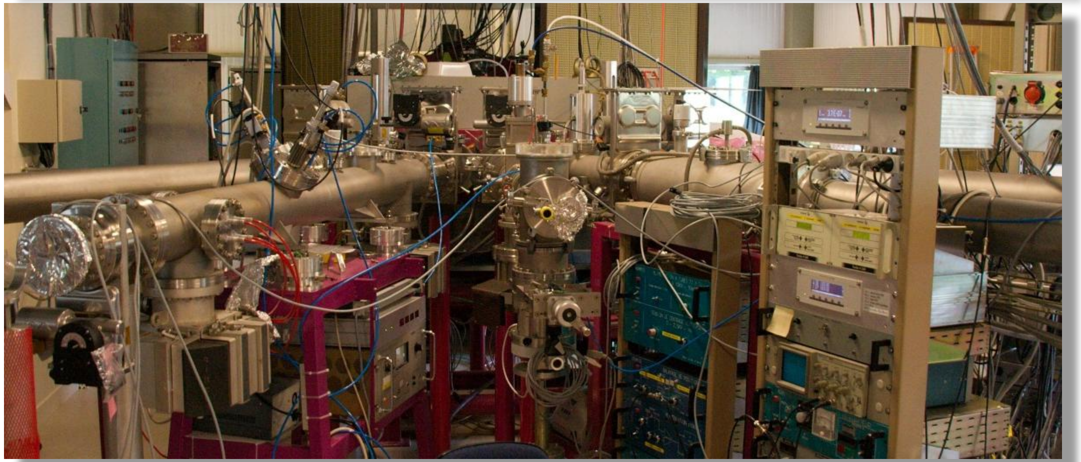


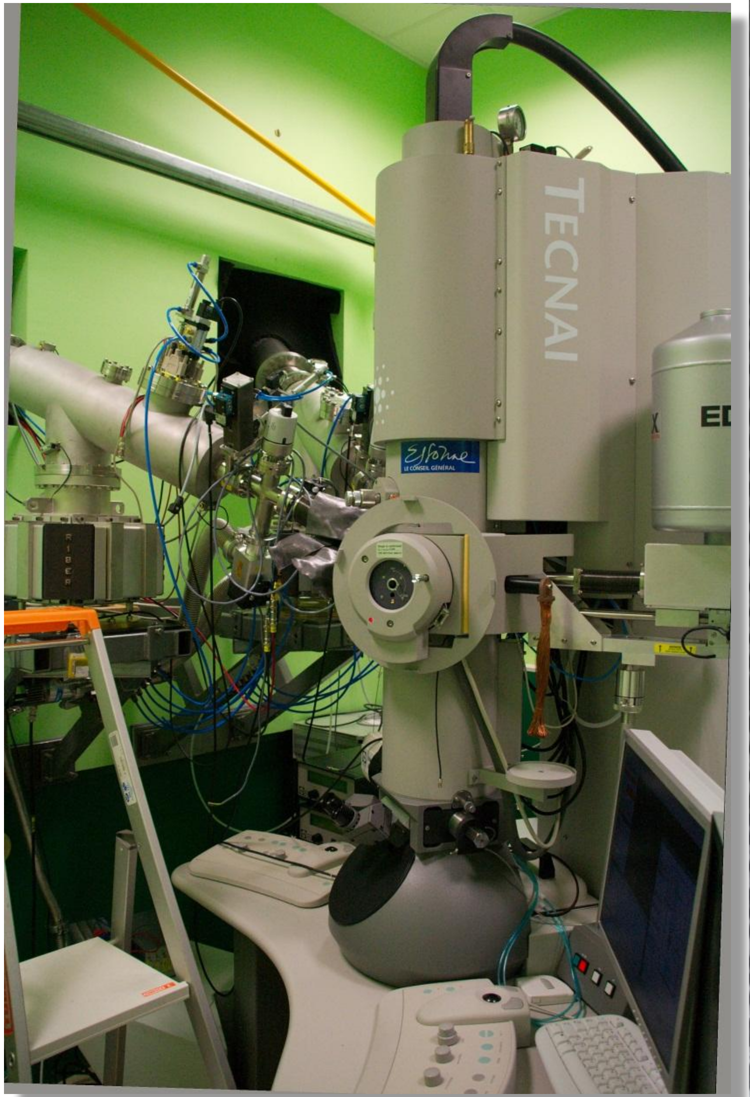
Selected Facilities: Hokkaido University, Sapporo, Japan

Specifications	
TEM	JEOL JEM-ARM1300
e-Beam Accelerating Voltage	400 to 1300 kV
400 kV Ion Beam	Fe, Ti, Ni or Ag Typical flux $2{\times}10^{10}$ ions.cm^{-2}.s^{-1}
300 kV Ion Beam	H, He, Ne or Ar Typical flux $4{\times}10^{10}$ ions.cm^{-2}.s^{-1}
Angle between Ion and Electron Beams	44°

Selected Facilities: JANNuS, Orsay, France

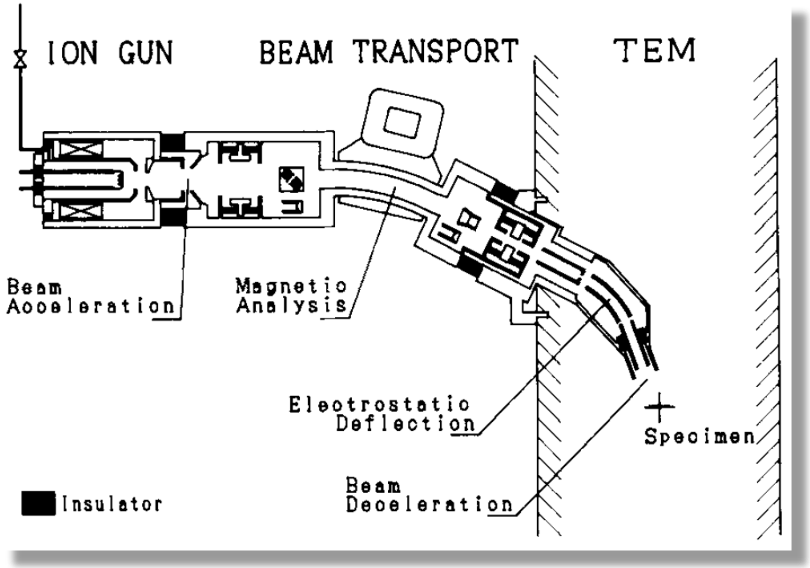
Specifications	
TEM	FEI Tecnai-200
e-Beam Accelerating Voltage	200 kV
IRMA	190 kV He to Pb
ARAMIS	2 MV H or He
Angle between Ion and Electron Beams	68°





Selected Facilities: Kyushu

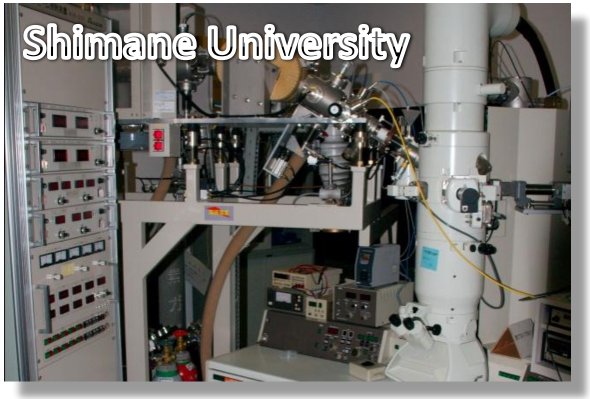
Specifications	
TEM	JEOL JEM-2000EX
e-Beam	200 kV
Ion Beam	0.1–10 kV
Ion Species	H, D, He
Angle between Beams	20°



J Nucl Mat **196–198** (1992) p1013

- Uses electrostatic deflection and deceleration inside the TEM column to achieve ion energies as low as 0.1 kV to explore, for example, the effects of plasmas on surfaces

Choice of TEM



- Interface ion beam: access and space above sample; or access through objective polegap; or able to be modified

- Space for ion beam current metering device(s)

- The actual magnifications required during actual irradiation are relatively low – although high resolution can be useful for further analysis

High Voltage Transmission Electron Microscope

- High voltage TEMs offer higher resolution, allow samples to be thicker and can also be used to introduce radiation damage

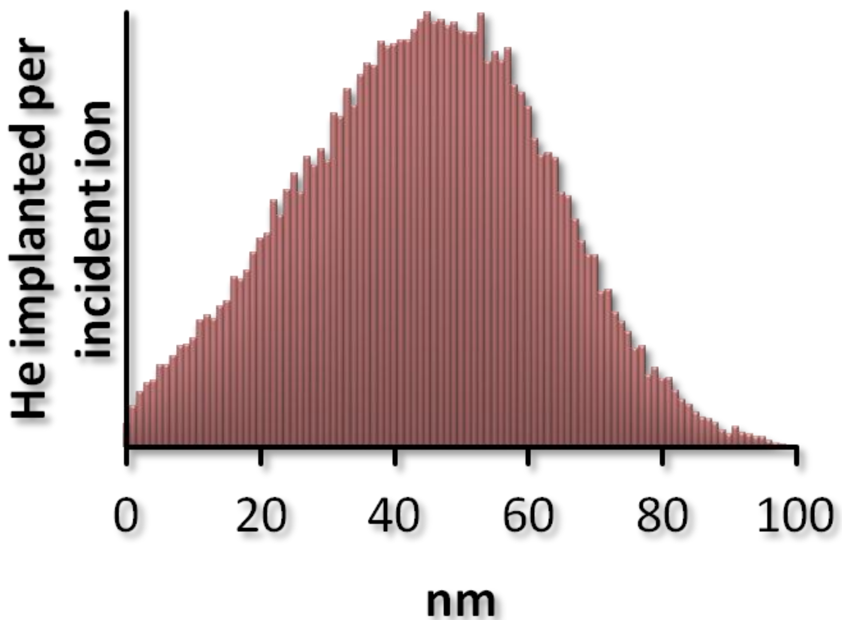
- Thicker samples reduce the effect of surfaces and accommodate the range of higher energy ions which can create 100 keV PKAs

- When operated above the threshold displacement energy for the sample, the electron beam can be used to create point defects either to simulate the effects of fission neutrons (displace individual atoms rather than cause cascades) or investigate effects of elevated point defect populations

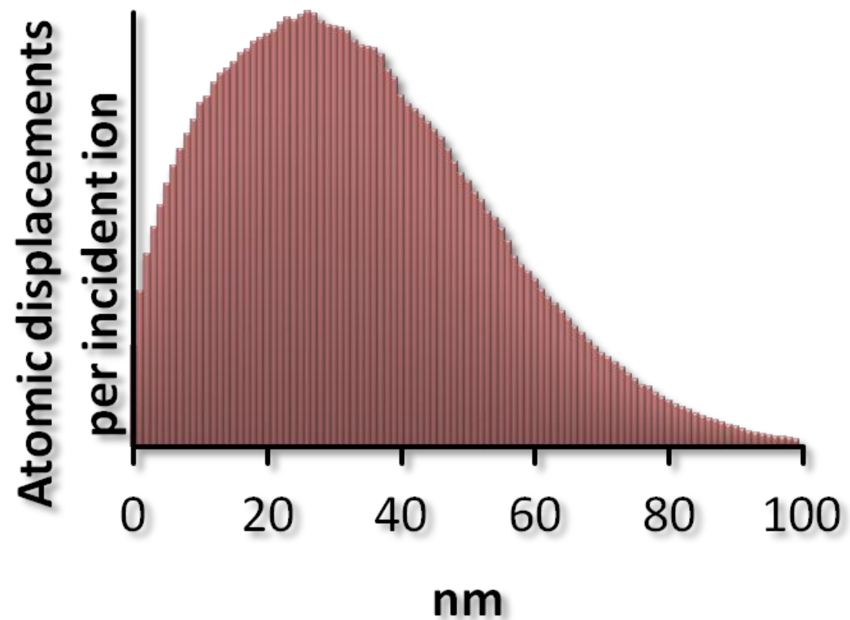
Choice of Ion Beam

- Electron transparent region of a TEM sample is generally up to 100 nm thick
- Ion species and energy must be chosen in order to achieve desired effects

6 kV He to SiC: R_p = 45 nm

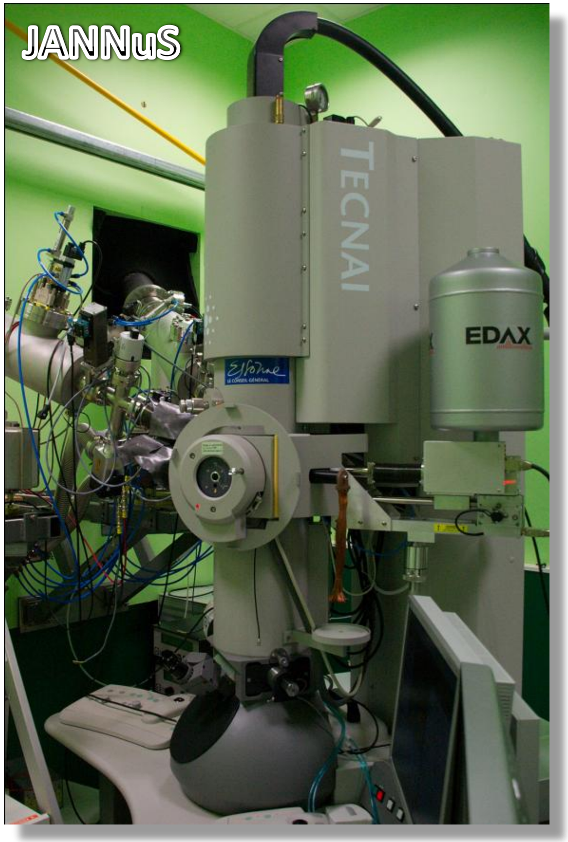


120 kV Fe to Fe: peak damage = 25 nm



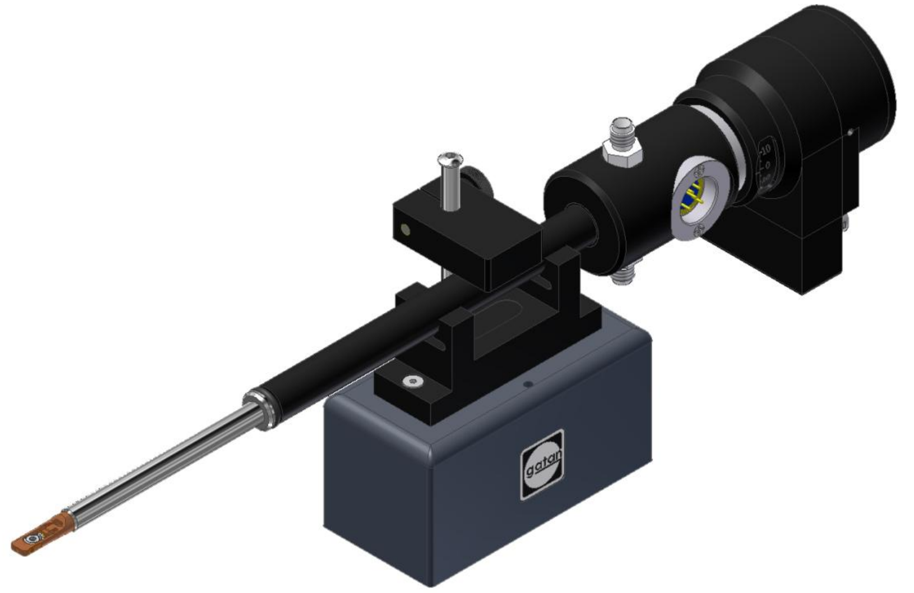
Multiple Ion Beams

- Two ion beamlines can be used to explore the synergistic effects of, for example, simultaneous He implantation and displacing irradiation or combined He and H bombardment
- There are four multibeam facilities currently in operation including the JANNuS system at CSNSM, Orsay, France
- This incorporates two beamlines with 190 kV and 2 MV accelerators

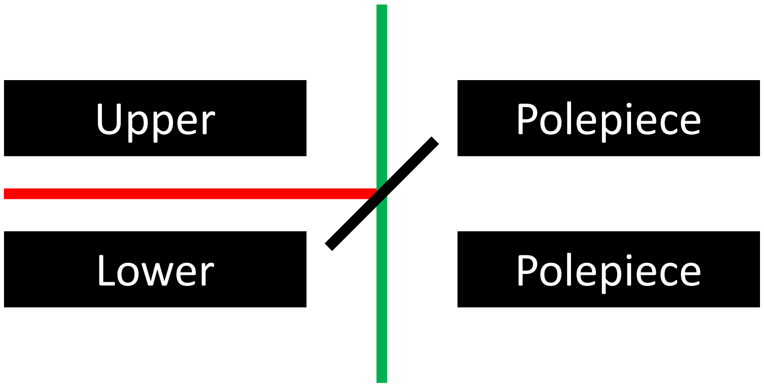


Optional Extras

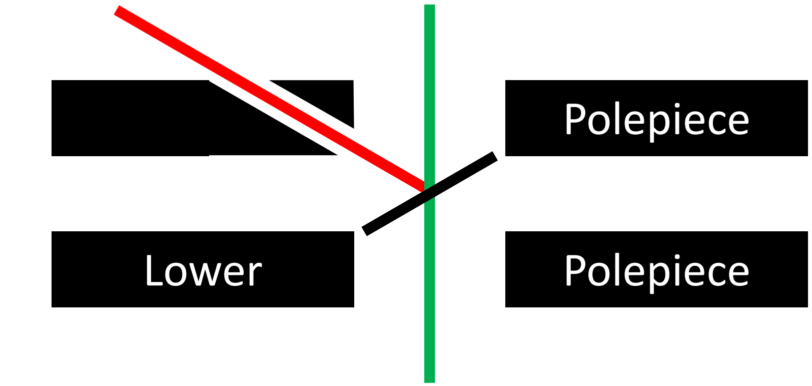
- Can employ normal range of sample manipulation, analysis and data capture
- TEM sample rods: heating, cooling, straining, environmental etc
- Analysis techniques: EDS, EELS, GIF etc
- CCD for digital capture of image and video



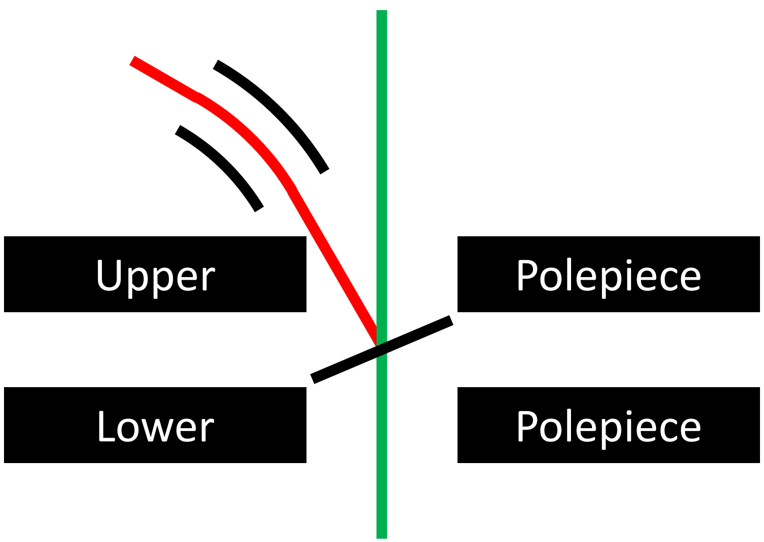
Interfacing of TEM and Ion Beam



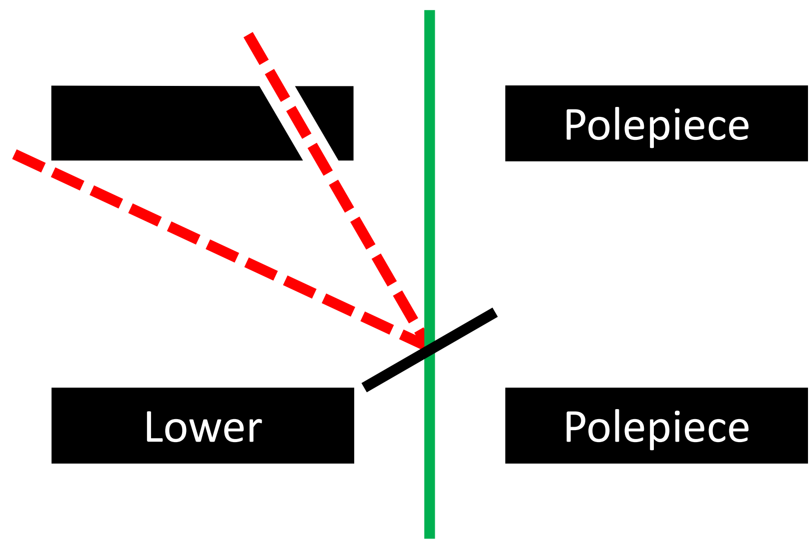
Side entry through polegap



Bore through upper objective polepiece



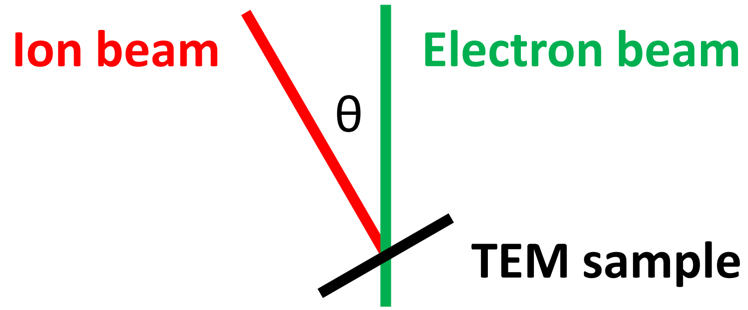
Electrostatic deflection inside TEM



Possibilities offered by larger polegap

Importance of Angle between Electron and Ion Beam

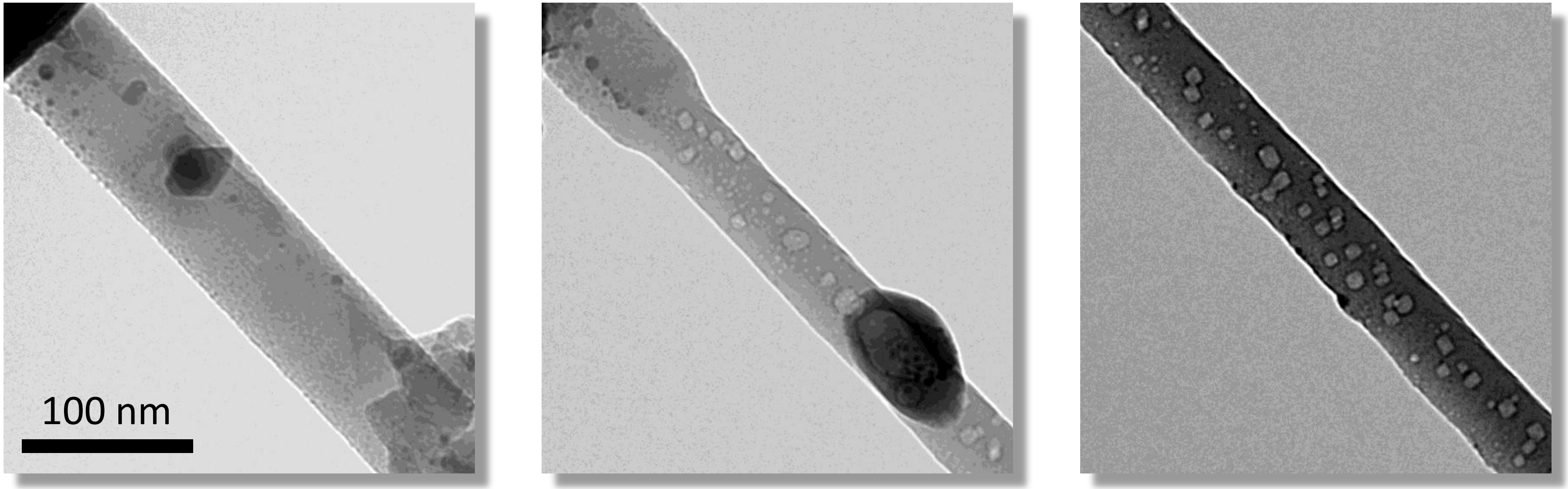
- Normal incidence vs glancing angle – usually desirable to be looking in direction close to the direction of irradiation



- Greater range of accessible sample tilt – if need particular crystallographic direction or orientation for analysis
- Larger usable thin area in sample – tilting away from normal incidence with e-beam means sample thickness increases
- Avoid shadowing effects – e.g. from TEM sample holder
- Implantation/damage profile is deeper for at a given energy

Shadowing Effects: a Cautionary Tale

- 6 keV He irradiation of Si nanowires at 500°C



- Shadowing effects can be very dramatic and can easily fool the best (!) of us
- The smaller θ is, the less likely shadowing is to occur!

So Much for the Past, What about the Future?

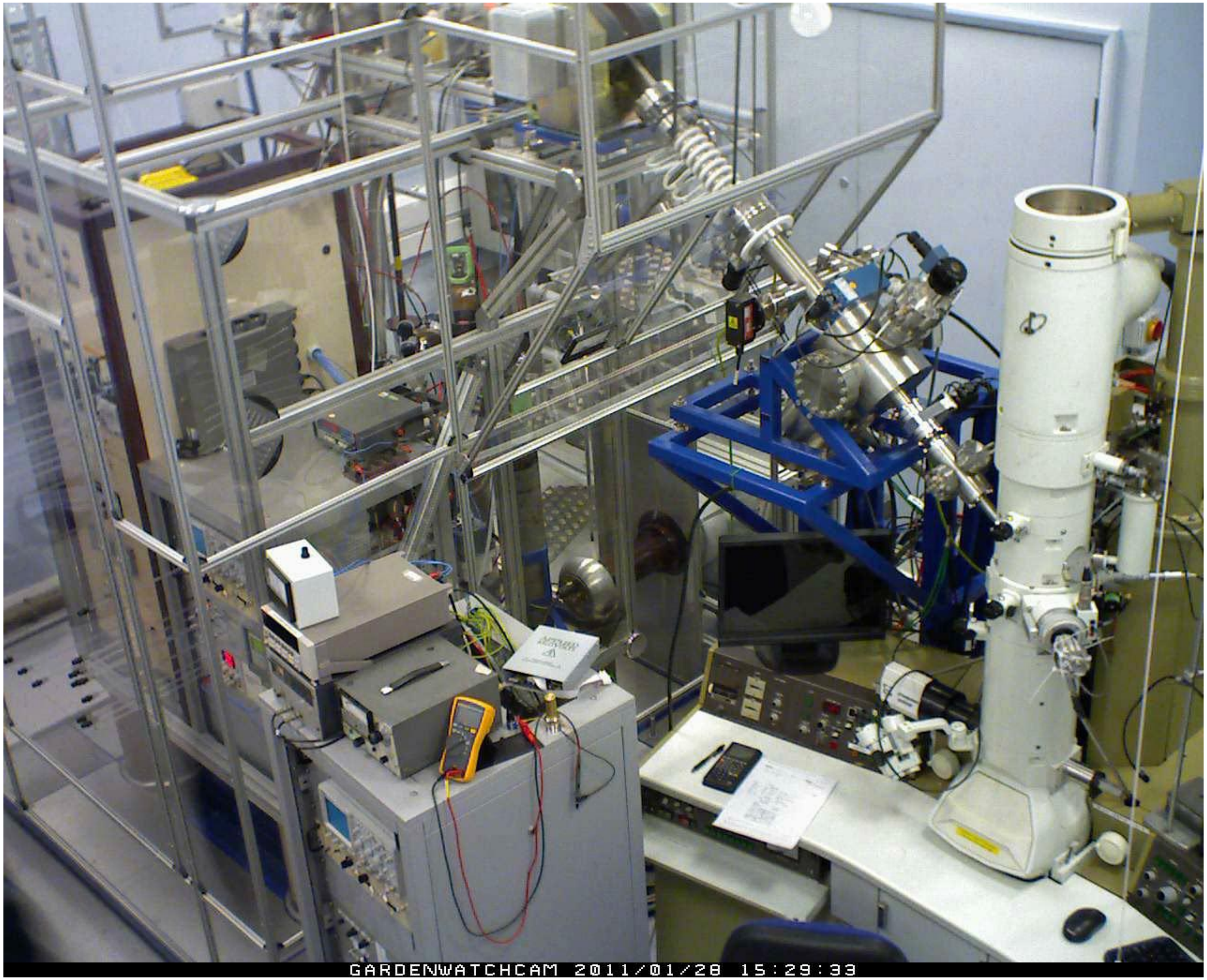
- New facility at Sandia National Laboratories
- Rumours of other facilities being planned in US have faded?
- New TEM sample rods offering ability to explore new conditions
- Aberration correction allows polegap to be opened for new geometries and reduction of θ.
- Will a nuclear revival drive second golden age?
- Our move from the University of Salford to the University of Huddersfield!

We are Currently Moving to Our New Laboratories

- New suite of purpose built laboratories at University of Huddersfield, West Yorkshire, England
- Everything has been dismantled and either has moved across or is moving today and tomorrow
- Used 1 mile of cling wrap to package everything up!




How to Disassemble a TEM with in situ Ion Irradiation



Second Workshop on the Use of In Situ TEM / Ion Accelerator Techniques
Albuquerque, New Mexico, USA, 6th June 2011

How to Reassemble a TEM with in situ Ion Irradiation

Coming soon …

(hopefully)

University of
HUDDERSFIELD

Thank you

j.a.hinks@hud.ac.uk