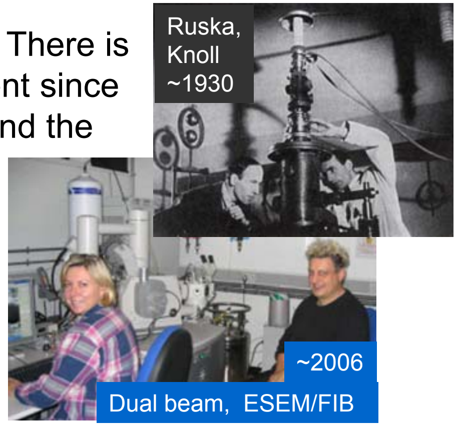
Frontiers of Experimental Condensed Matter Physics

- Part C, Atomic scale measurements: Part II: Electron Microscopy
  - Background and instrumentation
    - Scanning: SEM
    - Transmission:
      - TEM
      - STEM
  - Imaging and Contrast
    - Optics, Resolution limits
    - Scattering mechanisms
  - Variable pressure SEM
  - Analytical microscopy
    - EELS
    - EDX
  - Recent developments
    - Atomic resolution: aberration correction
    - 3-D microscopy

General ref: Williams and Carter "Transmission electron microscopy", Plenum Press (1996) – a comprehensive source used for many figures here.

Background

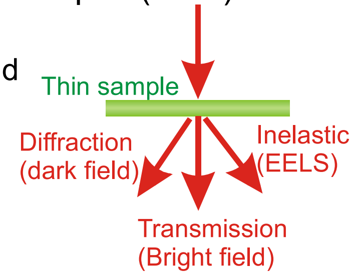
- History
  - After optical microscopy, electron microscopy is the most established microscopic method. There is ~70 years development since the first microscope and the subject is complex and detailed.



- Methods:
  - Different approaches arise from the many scattering and excitation processes.
    - Elastic scattering and diffraction – mainly in forward direction.
    - Inelastic scattering
      - single excitations: in the forward direction
      - multiple scattering: in all directions
    - Secondary electrons –created in the sample by excitation from the primary beam
    - Photons arising from excitation by the primary beam

TEM / SEM

- Observation of forward scattering requires thin samples and hence instruments known as Transmission Electron Microscopes (TEM)
  - The elastically scattered electrons, are coherent and can be imaged directly.
  - TEMs have the higher resolution (cf SEMs below).



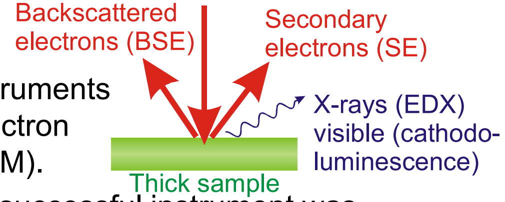
Thin sample

Diffraction (dark field)

Inelastic (EELS)

Transmission (Bright field)

- Observation of backscattered processes, from thicker samples.
  - The image is created by scanning a highly focussed probe across the sample and measuring the variation of the backscattered signal. Such instruments are Scanning Electron Microscopes (SEM).
    - The earliest successful instrument was constructed in the Engineering Department at Cambridge ~1952 (McMullan)
    - N.B. The STEM combines scanning and transmission methods.



Backscattered electrons (BSE)

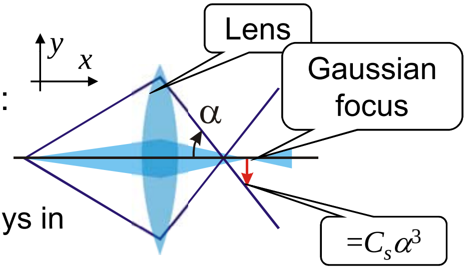
Secondary electrons (SE)

X-rays (EDX) visible (cathodo-luminescence)

Thick sample

Resolution

- **Factors affecting resolution:**
  - Probe size (which depends on the electron gun and electron energy), aberration of lenses, and diffraction at apertures.
  - Similar criteria limit both SEM and TEM instruments
- **Aberrations**
  - Spherical aberrations:
    - Gaussian focus is defined as the crossing point for rays in the limit $\alpha \to 0$.
    - As α increases, electron lenses “over-focus” and the crossing point moves to smaller x. The y-coordinate at the Gaussian focus varies like

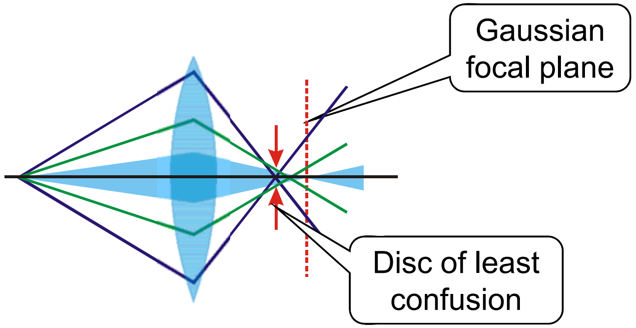


$$y(\alpha) \approx C_s\alpha^3 + O(\alpha^5)$$

NB This is part of a series expansion of the y co-ordinate in terms of α. The even order terms in α are zero by symmetry and the coefficient of the α^1 term is zero, since that defines the Gaussian focus.

Spherical aberration

- Note that the “best” focus occurs at the disc of least confusion, slightly away from the Gaussian focus.



- The spot size at the “best” focus is taken as

$$\delta_s \approx C_s \alpha^3$$

spherical aberration coefficient

half angle of beam

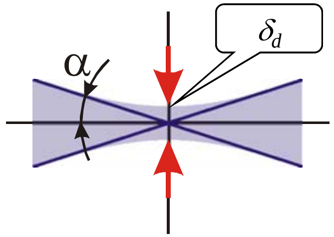
- There are other types of aberration (Coma, chromatic etc.); however, spherical aberration is usually the dominant factor.

Diffraction limit; Gun limit

Diffraction

- The ultimate focal spot is limited by diffraction. With a circular aperture, the diffraction pattern is an Airy disc (cf IB waves) of diameter.

$$\delta_d \approx 1.22\lambda / \alpha$$



Electron gun

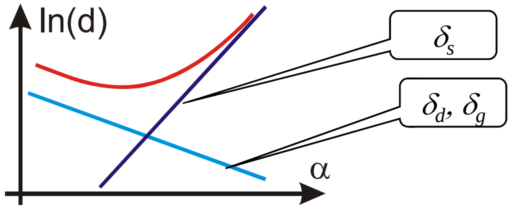
- The size of the electron probe is a compromise between the current, which determines the strength of the signal, and the half-angle of the beam.
- The key characteristic of the source is its brightness, β. i.e.. current emitted, per unit area, per unit solid angle.

$$\beta \approx \frac{i_e}{\pi\alpha^2}\frac{4}{\pi d_g^2} \Rightarrow d_g \approx \frac{2}{\pi}\sqrt{\frac{i_e}{\beta}}\frac{1}{\alpha}$$

i.e. for a given current the spot size is inversely proportional to the beam angle, α.

Resolution

- Adding diameters in quadrature $d^2 = \delta_s^2 + \delta_d^2 + \delta_g^2$
- The probe size (and resolution) is, ultimately limited by the lens aberrations.



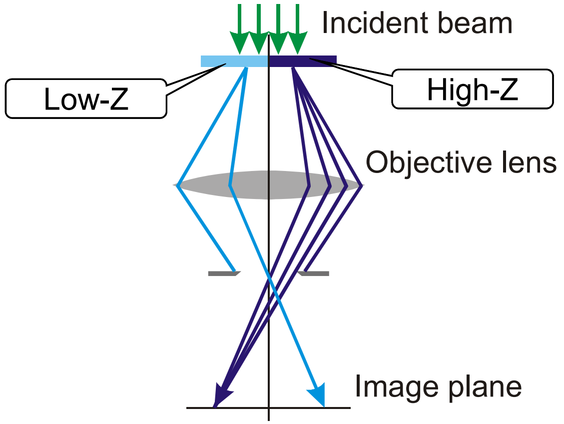
- Typical values:
  - SEM ~2-10 nm
  - TEM/STEM 0.1-1 nm
- Recent efforts to compensate for the spherical aberrations have lead to improvements. best resolution to date
    - SuperSTEM resolution <0.08 nm. See later

Contrast mechanisms

- There are many contrast mechanisms. Here we mention a few.

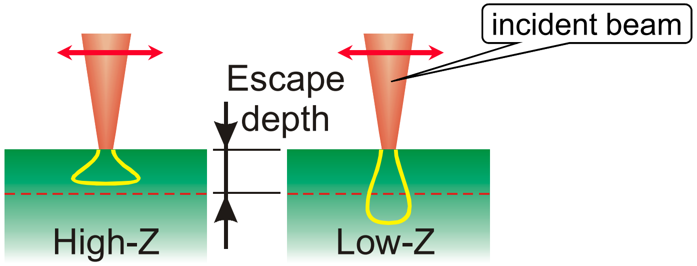
Mass-thickness and Z-contrast

- Recall that electrons scatter from other charges. Rutherford scattering from nuclear charge and core electrons will be greater for high-Z atoms (heavy materials), than for low-Z atoms (light materials).

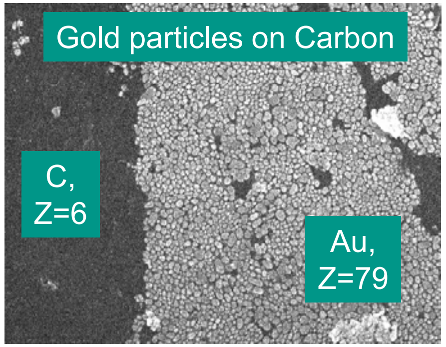


- Imaging (TEM) with the elastically scattered electrons leads to a Z-contrast image.

- A similar effect gives contrast with thick samples, i.e. in SEM. Sometimes called stopping-power contrast.

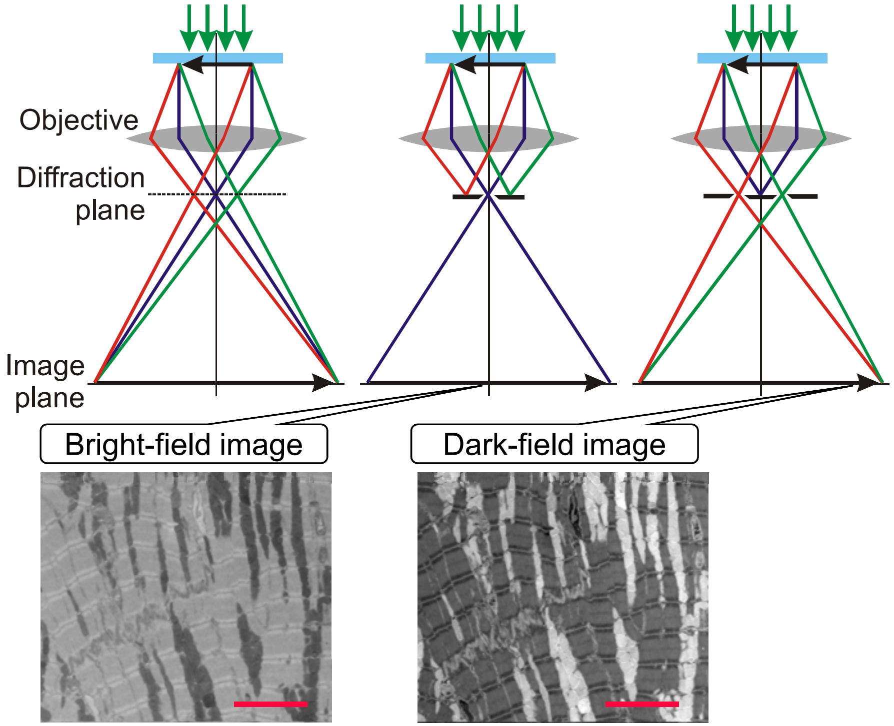


- Here, the stronger scattering from high-Z materials creates secondary electrons closer to the surface. They have an increased change of escape and hence appear brighter.



Diffraction contrast

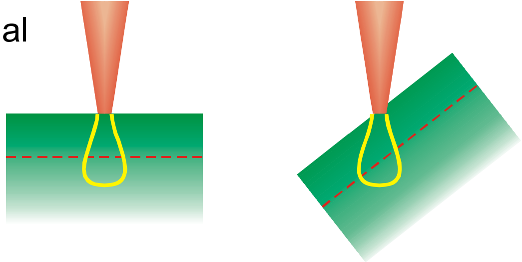
- Apertures located in the diffraction plane can select specific diffraction channels giving bright-field and dark-field imaging, as in optical microscopy.

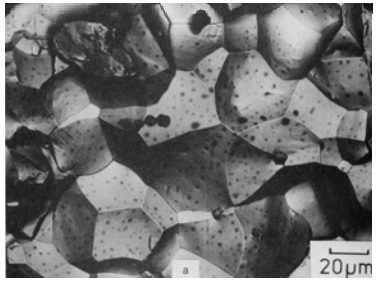


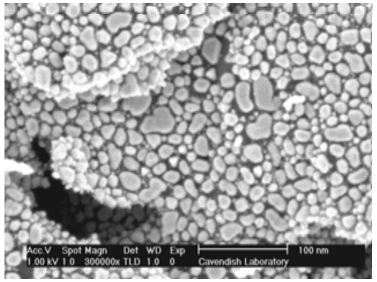
Cardiac Mice cells, fixed in glutaraldehyde, and osmium tetroxide, bulk stained in uranyl acetate,. Scale bar is 4mm

Topographic contrast

- The number of secondary electrons escaping in an SEM depends on the topography. The argument is similar to that for stopping-power (Z-contrast).
- Secondaries are created in a small volume, close to the surface, and the number that lie within the escape depth determine the brightness of the secondary electron image.
- Comparing normal and grazing incidence illustrates the point. A greater part of the secondary volume lies within the escape depth away from normal incidence.

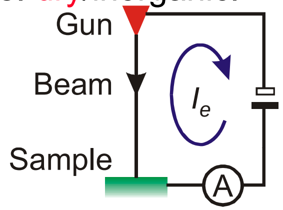




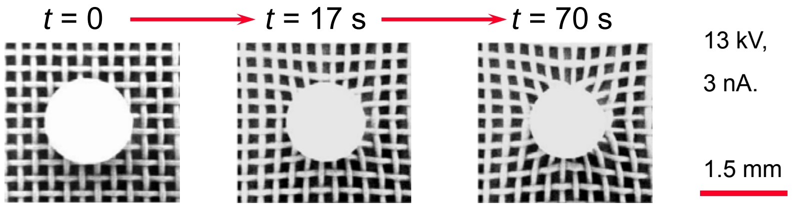


Some problems

- The microscope must be in a vacuum to transport electrons.
- Samples, in conventional instruments, must be compatible with the vacuum. i.e. dry/inorganic.
- We also require conducting samples since the current deposited in the sample must be part of a complete circuit.



- Figure, below, shows image distortion when an insulating, alumina sphere, Al_2O_3, becomes charged by the beam, so that strong electrostatic field develop, in time, around the sphere.



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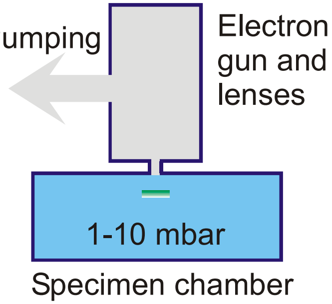
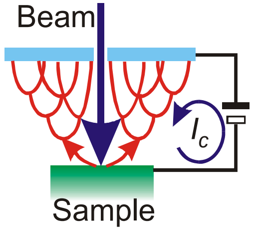
- Environmental chambers, giving higher pressure around the sample, have recently allow the observation of wet and non-conducting samples.

Variable pressure SEM/ESEM

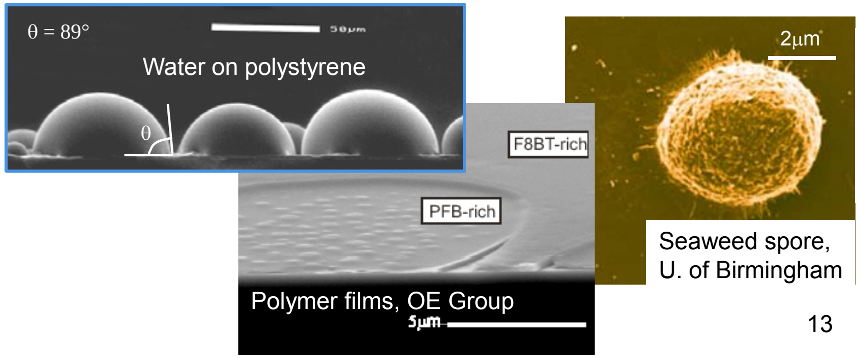
- At higher pressures:
  - Charge accumulation at insulating surfaces is reduced through neutralisation with ions in the gas (i.e. return path for the current is through the gas).





- There is cascade amplification of the secondary electrons.
- An environment saturated with water vapour aids all the above

- The Variable Pressure SEM (also known as Environmental ESEM) allows imaging of a variety of biological, insulating and soft-matter systems.



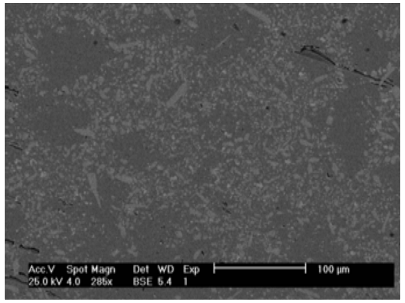
Water on polystyrene

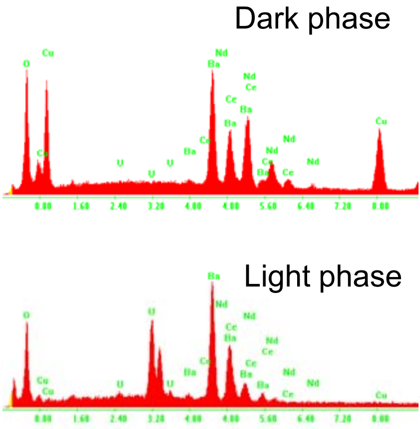
Polymer films, OE Group

Seaweed spore, U. of Birmingham

Analytic microscopy

- Microanalysis
  - So far we have discussed images derived from elastically scattered and secondary electrons. Other signals give spectroscopic information:
- X-ray emission:
  - Measurement of the energy of characteristic x-rays emitted by a sample is a standard feture of many SEM/TEM machines.
  - Illustration is from a sample of a doped superconductor, YBCO. The “light” phase and “dark” phase regions, seen in the left image, have different chemical constituents (see panels on the right)



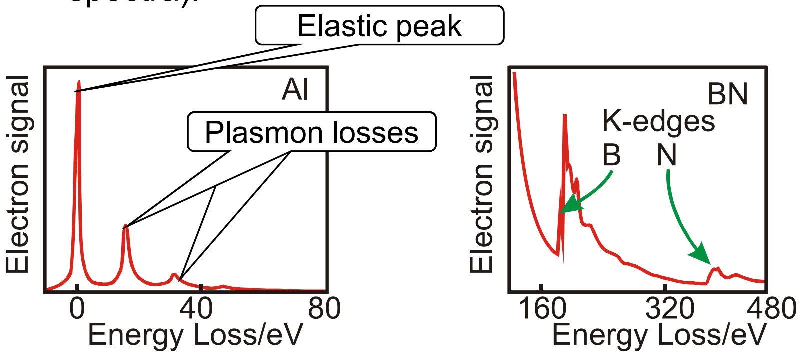


Electron energy loss

- Electron energy loss (EELS) is an alternative spectroscopic tool.
  - In TEM, measurements of the energy of the electron, after passing through the sample, indicate the inelastic scattering processes.

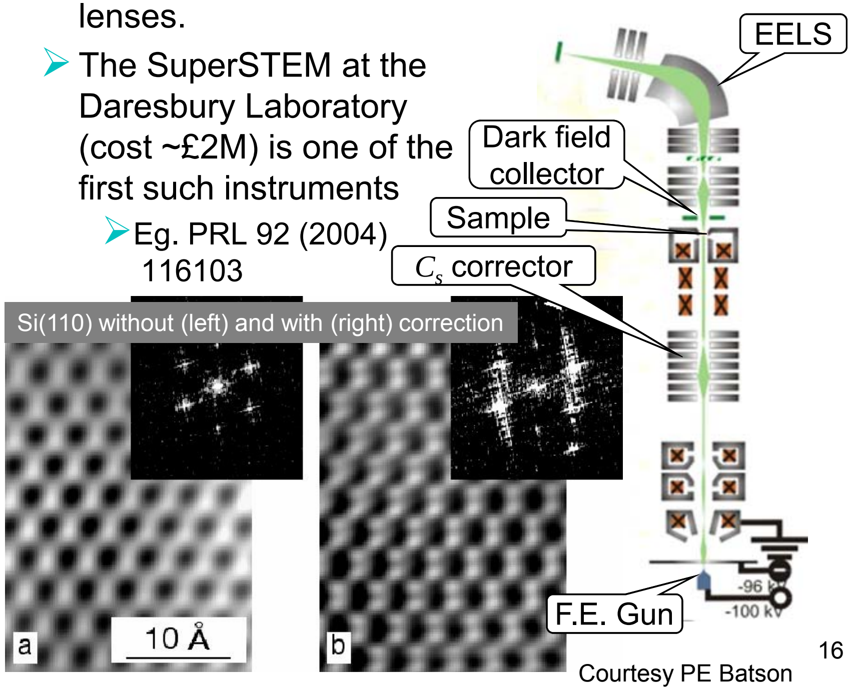
Process	Energy Loss (eV)	θ_E (mrad)
Phonons	~0.02	5-15
Plasmons	5-25	<-0.1
Inter/Intra-band transitions	5-25	5-10
Inner-shell ionization	~10-1000	1-5

  - Plasmon losses dominate in metallic systems. At higher energies core excitations occur (c.f. EXAFS spectra).



Recent developments I

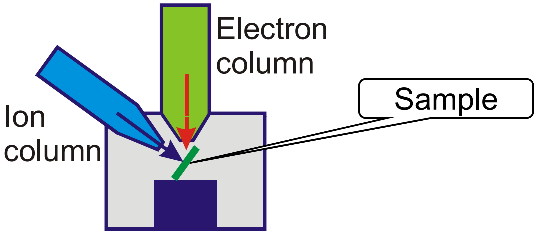
- Aberration corrected imaging, SuperSTEM
  - The idea is to compensate for positive spherical aberration, C_s>0, using a lens with a negative coefficient , C_s<0. One cannot do this with a cylindrically symmetric lens since all known systems have C_s>0. It can, however, be done with combinations of multipole (quadrupole/octopole) lenses.
  - The SuperSTEM at the Daresbury Laboratory (cost ~£2M) is one of the first such instruments
    - Eg. PRL 92 (2004) 116103



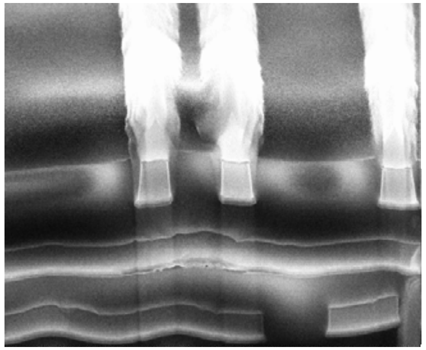
Courtesy PE Batson

Recent developments II

- 3-D imaging: Dual beam FIB/SEM
  - Focussed Ion Beam (FIB) selectively erodes the sample while SEM records images. From the 2-D images a 3-D image can be reconstructed



2-D images



Courtesy FEI

3-D reconstruction