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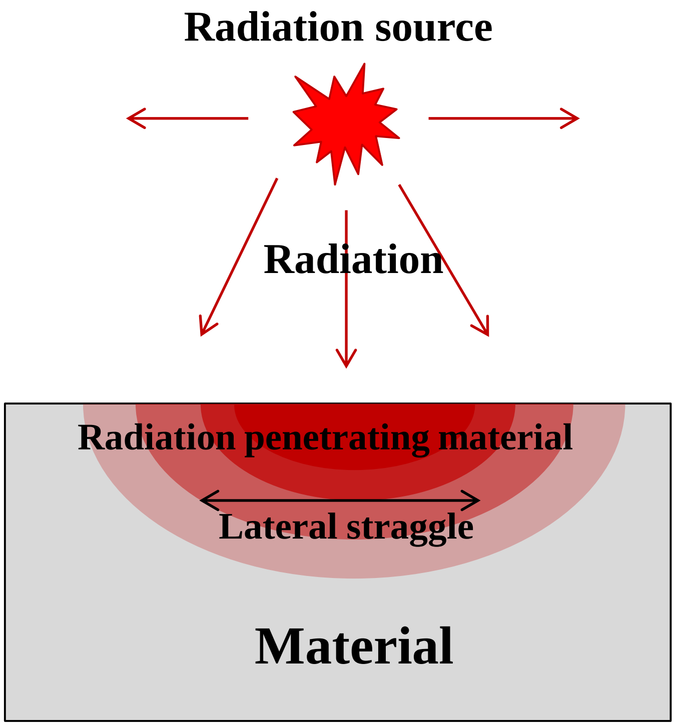
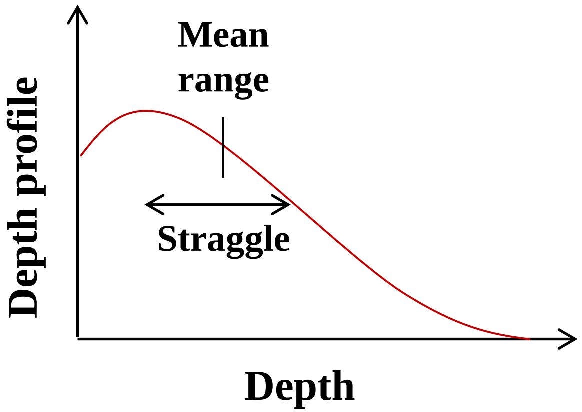
Radiation Damage in Materials

2. Overview of penetration mechanisms of energetic particles in matter

2.1. Penetration concepts

- For radiation penetrating matter (below the top atom layer) one can define:
- **Mean depth ≈ Mean range** (*medeldjup, medelräckvidd, keskisyvyys, keskikantama*)
- **Depth profile** (*djupprofil, syvyysjakauma*): concentration distribution of implanted particles
- **Straggling** (*spridning, leveys?*): spread of depth profile, usually defined as the standard deviation of the mean
- **Lateral straggling** (*lateral spridning, lateraalinen leveys*): spread in lateral direction





Mathematical expressions

- These can be calculated using the usual mathematical expressions for probability distributions
- A depth profile is in essence a probability distribution
- Assume the depth profile is known as $c(z)$, and surface is at $z=0$. $c(z)$ obviously has to be > 0 for all z
- Then:
  - Total concentration of implanted material: $C = \int_0^\infty c(z)dz$
  - Mean range $\bar{R} = \frac{1}{C} \int_0^\infty zc(z)dz$
  - Straggling $\sigma = \frac{1}{C} \int_0^\infty (z - \bar{R})^2 \, c(z)dz$
- Also any other probability distribution concept can be used, e.f. median, skewness (*snedhet/vinous*), kurtosis (*kurtosis, huipukkuus*), full-width-half-maximum, etc.

Other terminology

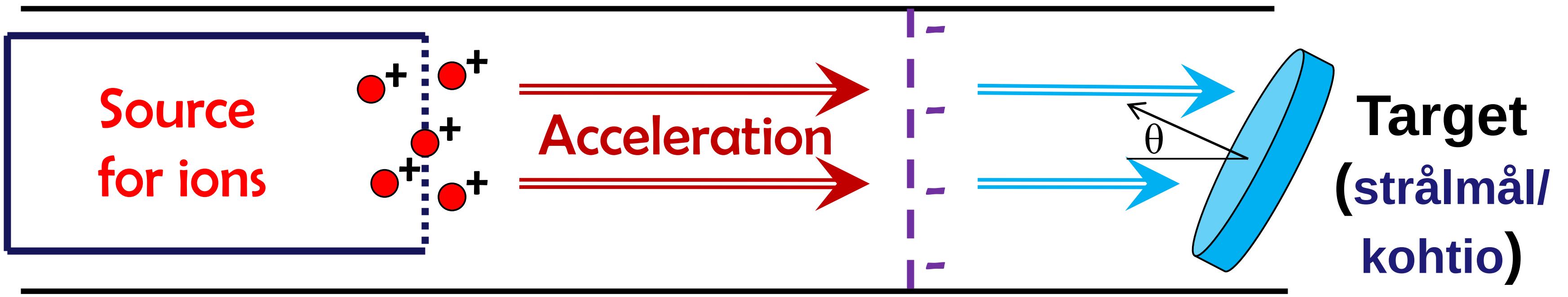
- **Reflection** (*reflektion / sironta*): particle does not penetrate at all but is reflected
  - Or actually may penetrate to some depth but then is reflected back
- **Deposition** (*deposition / depositio*): particle stays in the substrate
  - This can be both deposition above the surface or deeper in
- **Sticking** (*stickande/klibbning / kiinnittyminen*): particle sticks on top of surface
- **Sputtering** (*sputring eller förstoftning / sputrautminen tai roiskuminen*): particles from inside material are kicked out due to the radiation process

How are these quantified?

- **Reflection** (*reflektion / sironta*): measured with reflection coefficient, i.e. fraction of particles reflected (*reflektionskoefficient / sirontatekijä)*
- **Deposition** (*deposition / depositio*): measured with fraction of deposited particles
- **Sticking** (*stickande/klibbning / kiinnittyminen*): measured with sticking coefficient (*stickningskoefficient / kiinnittymistekijä*)
- **Sputtering** (*sputring eller förstoftning / sputrautminen tai roiskuminen*): measured with **sputtering yield** (*koefficient/utbyte/tuotto*) = number of outcoming atoms / number of incoming ions

Radiation exposure conditions

- Radiation impacting on a material can be monoenergetic or have an energy spread
- It can come at a well-defined **incoming angle** (*inkomstvinkel / sisääntulokulma*) or with an angular distribution
  - If it is a beam, it usually has a pretty precise incoming angle
  - Beam spread: in reality even beams always have some spread, can be e.g. ±0.5º. This is often ignored as insignificant.
- Accelerators typically have a well-defined energy, beam and angle θ. Simplified picture:



2.2 Overview of penetration

- A key aspect for the penetration of particles with matter is whether they are neutral or charged
- Neutral particles do not interact with the electromagnetic force, and hence interact weakly with matter -> big ranges
  - MeV Neutrons: meters!
    - Scattering depends on mass difference, most efficient scattering by same mass particles = protons
    - Hence water good to slow down neutrons in nuclear reactors
  - Gammas: centimeters
    - E.g. 1 MeV gamma: 8 mm in lead, 44 mm in concrete

http://www.ndt-ed.org/EducationResources/HighSchool/Radiography/penetrationdepth.htm

Overview of penetration

- Charged particles interact strongly: likely lots of scattering, only small range
  - Electrons: E.g. 1 MeV electrons in Copper, adjacent equation by Potts gives 350 µm

$$x\,(\mu m) = \frac{0.1\, E_o^{1.5}}{\rho}$$

where E_o = accelerating voltage (keV), and ρ = density (g/cm^3)

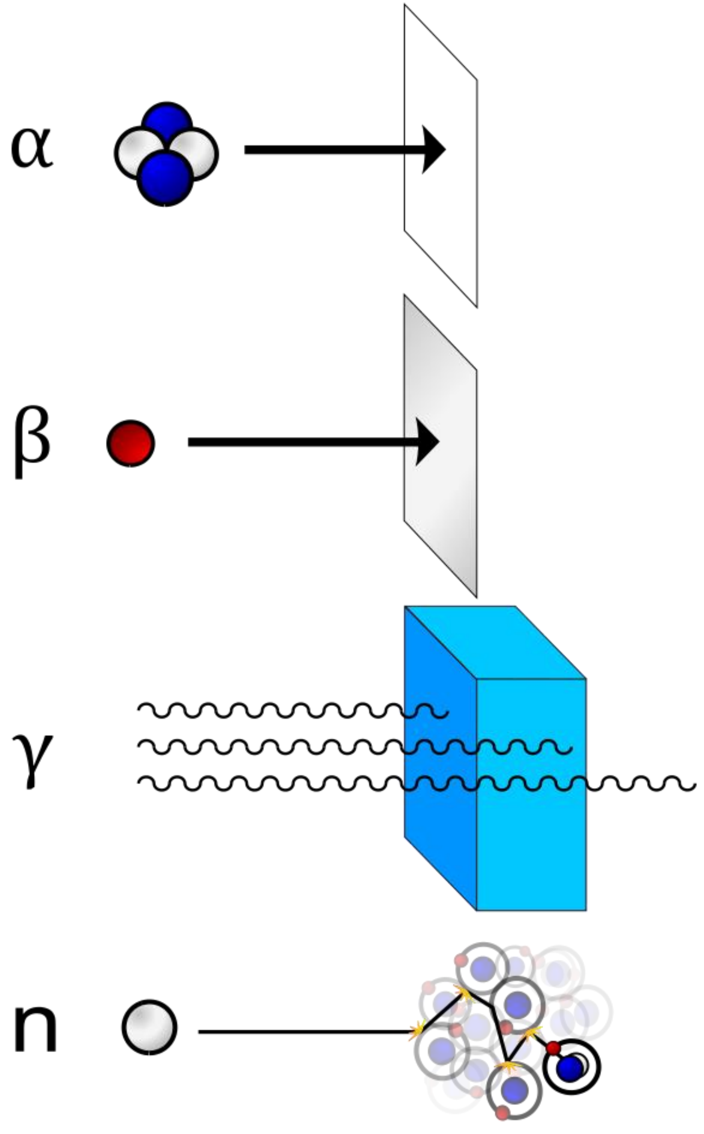
http://www4.nau.edu/microanalysis/Microprobe/Interact-Volume.html

  - Ions:
    - 1 MeV protons in Cu: 380 nm,
    - 1 MeV Au ions in Cu: 96 nm

    [Numbers from SRIM]

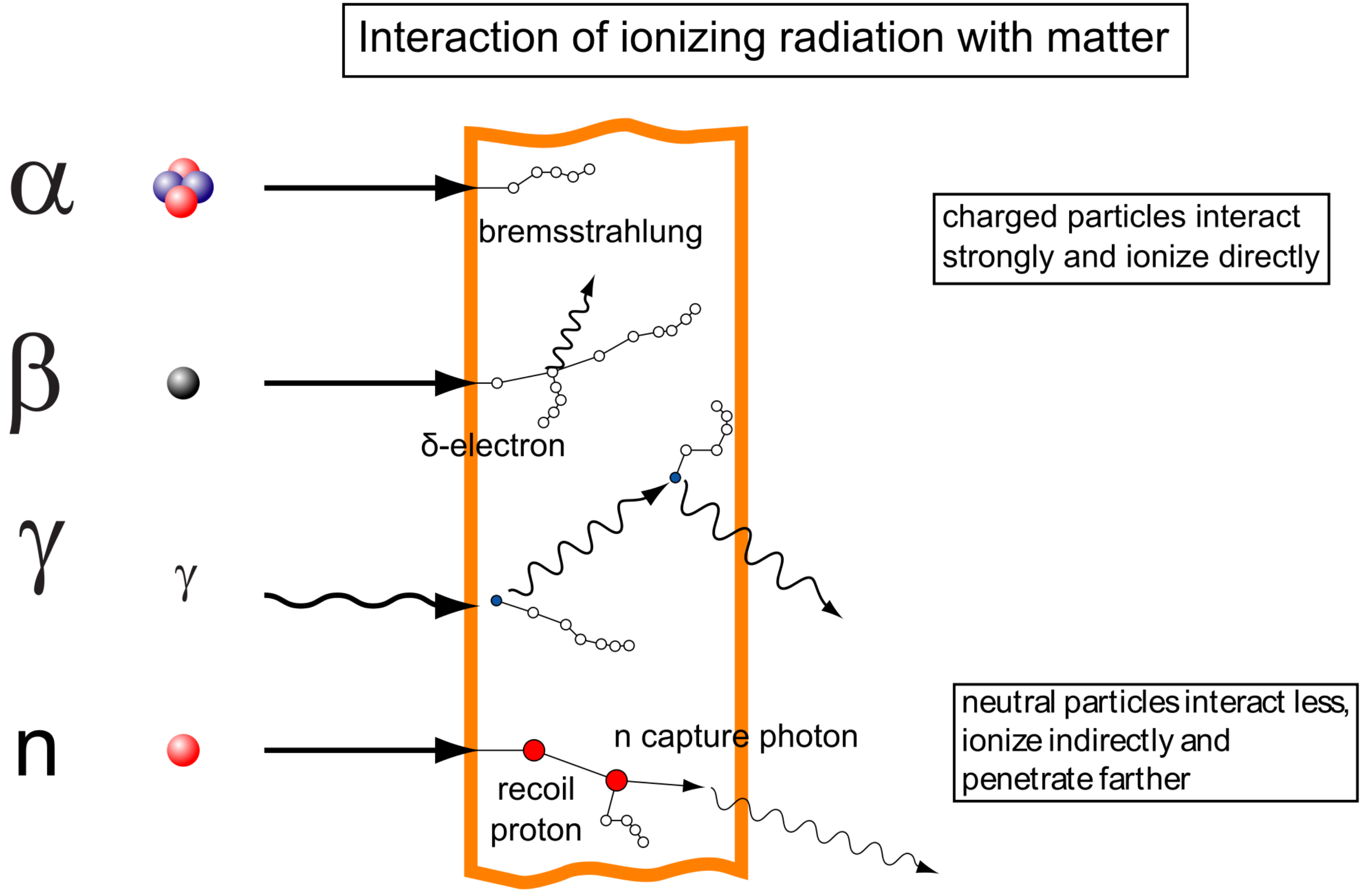
Overview of penetration

- Illustration [wikipedia]:
  - A sheet of paper is enough to stop alphas
  - An aluminum sheet enough to stop electrons
  - Gammas need more
  - Neutrons need meters of material



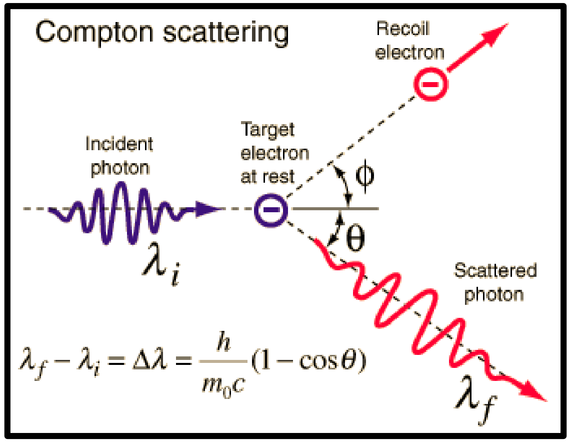
[wikipedia: ionizing radiation]

Illustration 2



[wikipedia: ionizing radiation]

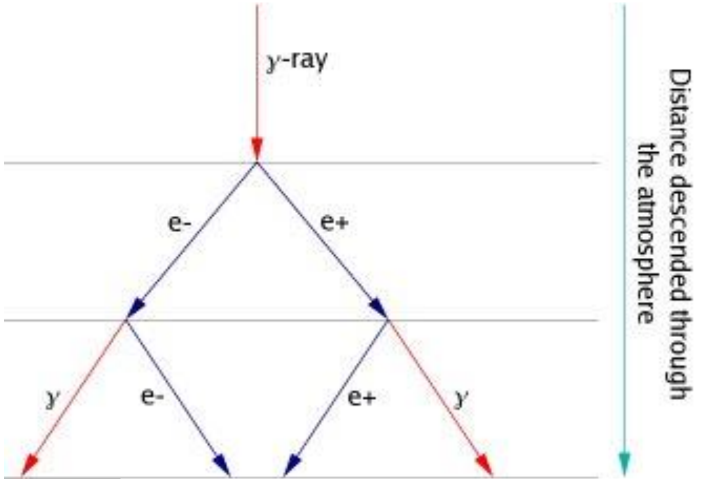
2.3 Photons

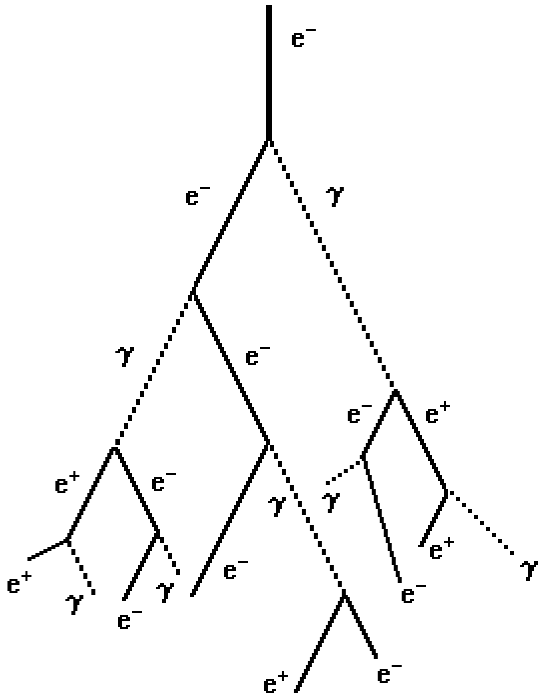


- High-energy photons penetrating matter when slowing down can ionize electrons with the photoelectric effect and Compton scattering
  - These in turn can excite atoms, and when these decay, they create additional photons
    - These in turn can create additional ionized electrons
      - These can crate more photons…
- The end result is known as **electron-gamma cascades** (*elektron-gamma kaskader, elektroni-gamma-kaskadi/ryöppy*)
  - If the energy is higher than > 1022 keV, it can also produce electron-positron pairs, known as pair production
    - Limit due to rest mass of electron, 511 keV

Electron-gamma cascades

- From the explanation above, it is clear that both electrons and gammas can induce electron-gamma-cascades
- Hence the penetration of both gammas and electrons can and are often treated with the same codes
- Concept works down to energies ~ 100 eV – 1 keV
  - Below that, e's and γ's do no longer behave particle-like but quantum wave functions => very difficult to model and understand

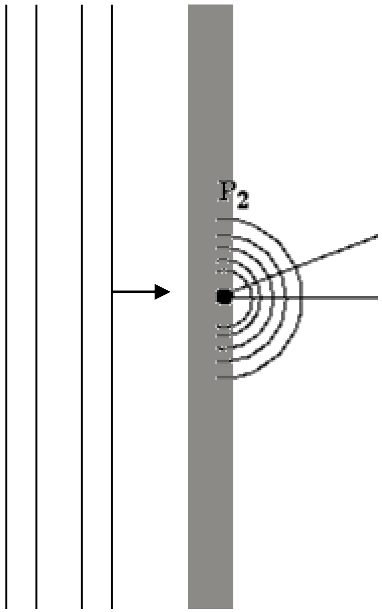


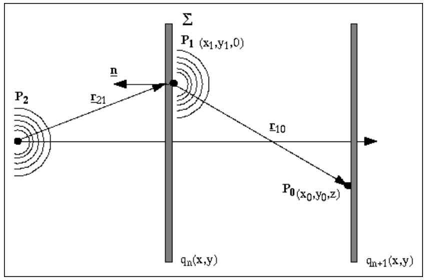


2.4. Electrons

1. See above! High-energy single electron penetration is similar to gammas
2. But a beam of monoenergetic electrons at energies around 100 keV are the basis of transmission electron microscopy (TEM) operation, and in that case can be treated in a wave mechanics way.

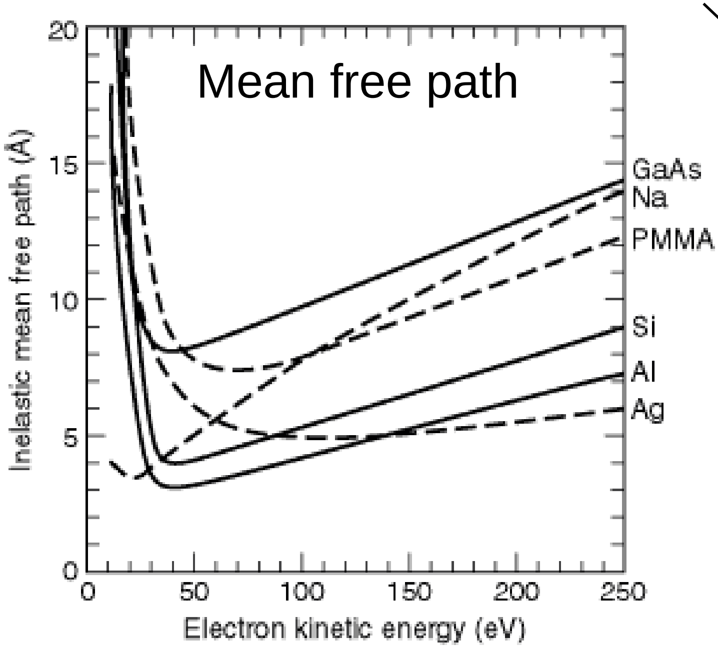
- TEM operation can be well understood and modelled with waves, which scatter from the electrons of atoms as scattering centers
- There is no paradox here, the issue just reflects the basic quantum mechanical particle-wave duality, i.e. that depending on aim, matter can be either treated as particles or waves

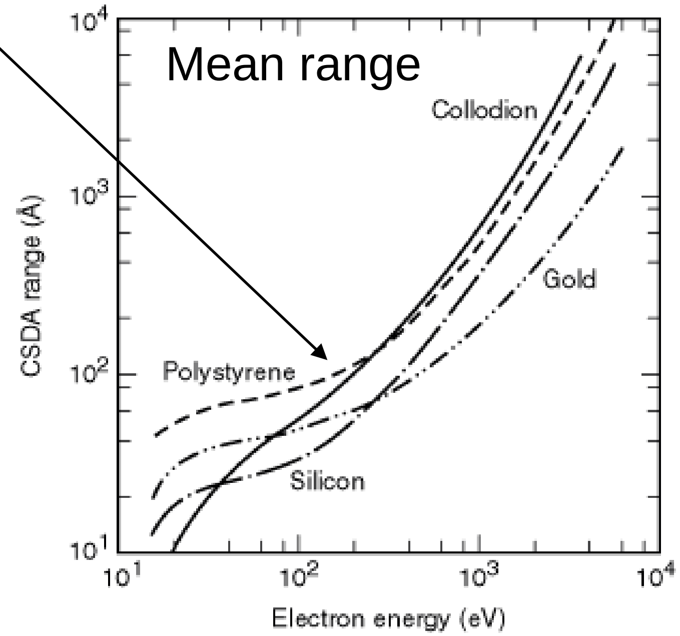




Electrons: low energies

- At very low energies, the electron scattering mean free path in materials has a slightly unexpected behavior: it has a **minimum** around roughly 50 eV due to details of interaction of the electron with the electronic structure
  - Below 50 eV the mean free path increases again
  - Due to this, also the electron mean range as a function of energy has a somewhat nonobvious shape





[https://xdb.lbl.gov/Section3/Sec_3-2.html]

Electron – nucleus knock-ons

- Electrons can also with a small but not insignificant probability hit nuclei
- This (usually) is an elastic binary collision governed by classical kinematics
- Often (e.g. in TEM's) main source of radiation damage
- Basic classical binary collision equations give the maximum energy transfer to an atom of mass m_{atom} as

$$E_{max,atom} = \frac{4\, m_{electron} m_{atom}}{(m_{electron} + m_{atom})^{\,2}} E_{electron}$$

where $E_{electron}$ is the kinetic energy of the electron and $E_{max,atom}$ the maximum energy an atom can receive.

Electron – nucleus knock-ons: approximation

- Since the electron mass is always at least about 2000 times lower than the atom mass, one can approximate

$$m_{electron} + m_{atom} \approx \; m_{atom}$$

  and hence simplify the equation as:

$$E_{max,atom} \approx \frac{4\, m_{electron}}{m_{atom}} E_{electron}$$

- From this form, it is obvious that the atom receives only a small fraction of the electron energy!
  - E.g. for Si with m_{atom}= 28u, using $m_{electron}$= u/1823, one obtains $E_{max,atom} \approx \frac{4}{28*1823} E_{electron} = \frac{E_{electron}}{12760}$
  - In other words, to get recoil energies exceeding 10 eV, one needs electrons with energies > 100 keV

Electron – nucleus knock-ons

- However, for high electron energies one needs to use the relativistic kinematics version of this equation, which is

$$E_{max,atom} = \frac{2ME\ (E+2mc^2)}{(m+M)^2c^2+2ME}$$

  where the notation has been shortened with

  $m_{electron} = m,\ m_{atom} = M, E_{electron} = E$

- Of course the probability of a direct knock-on leading to the maximum energy transfer is very small, but this is still an important equation since it allows calculating the threshold displacement energy

McKinley-Feshbach equation

- Calculation of the full displacement probability is complicated, but the McKinley-Feshbach equations provide a good approximation [Phys. Rev. 74 (1948) 1759]
  - Useful reference on implementation: [Lucasson, Phys. Rev. 127 (1962) 485]
- Cross section is slightly complicated:

$$\sigma = \frac{4Z^2 E_{\mathrm{R}}^2}{m^2c^4}\left(\frac{T_{\max}}{T_{\mathrm{thr}}}\right)\pi a_0^2\left(\frac{1-\beta^2}{\beta^4}\right)\left\{1+2\pi\alpha\beta\left(\frac{T_{\mathrm{thr}}}{T_{\max}}\right)^{1/2} - \frac{T_{\mathrm{thr}}}{T_{\max}}\left[1+2\pi\alpha\beta+\left(\beta^2+\pi\alpha\beta\right)\ln\left(\frac{T_{\max}}{T_{\mathrm{thr}}}\right)\right]\right\} \quad (5)$$

where Z is the atomic number of the displaced atom, E_{R} the Rydberg energy (13.6 eV), α_0 the Bohr radius of the hydrogen atom (5.3×10^{-11} m), $\beta = v/c$, and $\alpha = Z/137$. It can be seen that σ increases with increasing Z, therefore heavier atoms should be displaced more easily; however, due to momentum conservation (equation (3)), the energy transfer is lower. At very high projectile energies ($E \gg E_{\mathrm{d}}$) a higher displacement rate can be expected for heavier atoms.

[Banhart, Rep. Prog. Phys. 62 (1999) 1181]

McKinley-Feshbach equation

- This is widely used still as sort of a standard, but it is good to remember it is approximate
- Right top is a comparison to experiments from the original paper

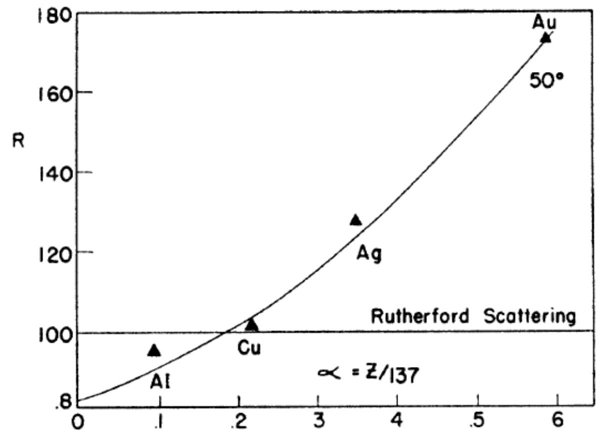


FIG. 5. Comparison of theory and experiment at electron energy of 2 Mev. The solid line is given by the theory. The triangles give the experimental points as obtained by Van de Graaff, Buechner *et al.*

- Right bottom: decrease of cross section with energy

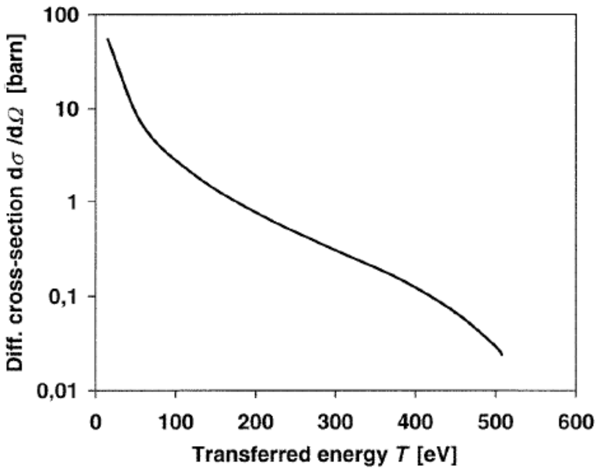


Figure 3. Differential cross section $d\sigma/d\Omega$ of the displacement of a carbon nucleus by an electron as a function of the energy T transferred to the nucleus ($E = 1$ MeV, $T_d = 10$ eV).

[Banhart, Rep. Prog. Phys. 62 (1999) 1181]

Comparison of electron interaction cross sections

- For carbon atoms:

Table 1. Cross sections σ for the interaction of electrons (100 keV) with an agglomerate of carbon atoms (Cosslett 1978).

Ionization	$\sigma = 2.0 \times 10^6$ barn
Plasmon excitation	1.5×10^6
X-ray emission (K-shell)	2.5×10^4
Displacement of an atom	1.8×10^2
Elastic collision (diffraction)	10^6

[Cosslett, 1978 *J. Microsc.* **113** 113–29]

- Hence the atom displacement cross section is indeed small, but it can be dominating for damage production, since in many materials ionization does not produce any damage

2.5 Neutrons

- Neutrons do not interact significantly with electrons, but mainly with atomic nuclei via the nuclear forces
- Since the radius of nuclei and the range of the nuclear forces is is of the order of fm (femtometers), the collision probability is very small => hence the long ranges of neutrons in matter
- Neutrons can interact with several different processes with the atoms, the most important of which are:
    1. Elastic scattering
    2. Inelastic scattering
    3. (n,2n) reactions
    4. (n,γ) reactions
    5. Nuclear fission

Elastic neutron scattering

- The elastic neutron scattering can be calculated with classical kinematics, similar to electrons
- But since the nature the interaction is different, it is not exactly the same equations
- Detailed calculation: Was book, section 1.1.1.

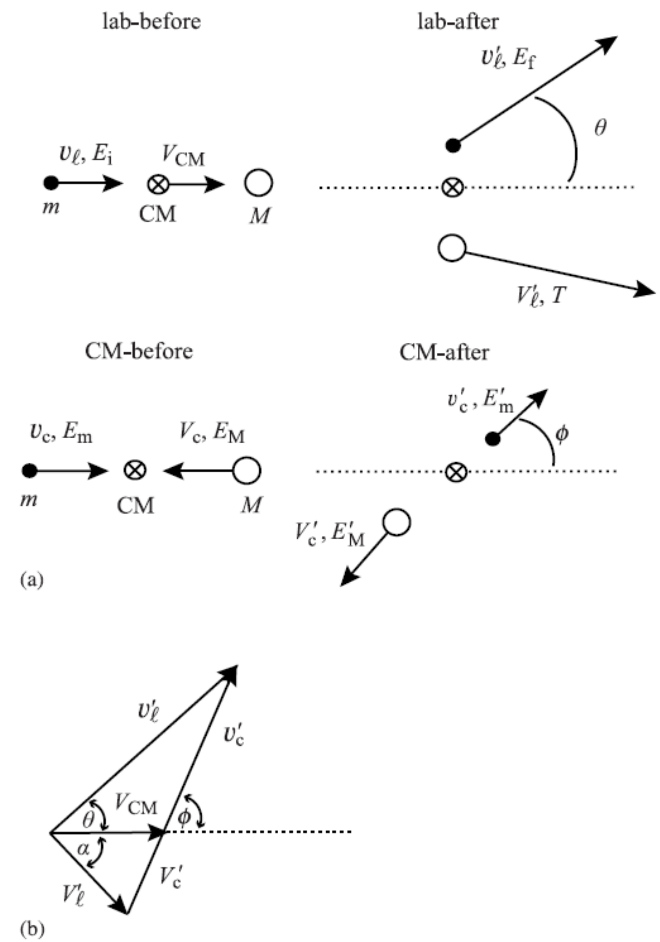


Fig. 1.1. Vector velocities (a) in the laboratory and center-of-mass (CM) systems, and (b) composite diagram relating velocities in the two systems

Elastic neutron cross sections

- The calculation gives

$$T = \frac{\gamma}{2} E_{\mathrm{i}} \left(1 - \cos\phi\right)$$

with

$$\gamma = \frac{4mM}{(M+m)^2}$$

where E_i is the initial energy of the neutron of mass m, T is the energy transferred to the atom of mass M, and ϕ is the scattering angle in center-of-mass coordinates (see picture on previous page)

Elastic neutron cross sections

- The angular dependence is as follows
- The maximum corresponds to a knock-on collision for backward scattering of the neutron, and is the same equation as given for maximum energy transfer for electrons

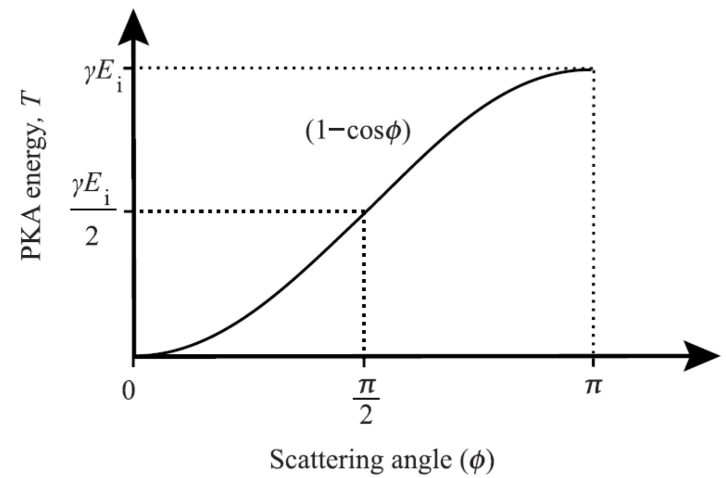


Fig. 1.2. Energy transfer as a function of center-of-mass scattering angle

- A further calculation (see Was sect. 1.1.1) gives as an example the following values for the average energy transfer in a collision:

$$T \approx \frac{\gamma E_i}{2}$$

$$\begin{aligned}
&1\,\mathrm{MeV}\ \mathrm{n}\ \mathrm{on}\ \mathrm{C}: && \gamma = 0.28 && \bar{T} = 0.14\,\mathrm{MeV} \\
&1\,\mathrm{MeV}\ \mathrm{n}\ \mathrm{on}\ \mathrm{Fe}: && \gamma = 0.069 && \bar{T} = 0.035\,\mathrm{MeV} \\
&1\,\mathrm{MeV}\ \mathrm{n}\ \mathrm{on}\ \mathrm{U}: && \gamma = 0.017 && \bar{T} = 0.009\,\mathrm{MeV}
\end{aligned}$$

Inelastic neutron scattering

- Inelastic neutron scattering becomes important for neutron energies above 1 MeV
- It means scattering of a neutron such that the kinetic energy is not preserved, but goes into a excitation of the (potential) energy of the nucleus
- Written as a nuclear reaction equation: $^{A}X(n,n')\,^{A}X^{*}$ where ^{A}X is the isotope (e.g. ^{14}N or ^{11}B) and the asterisk * denotes that the nucleus has become excited

(n,2n) reactions

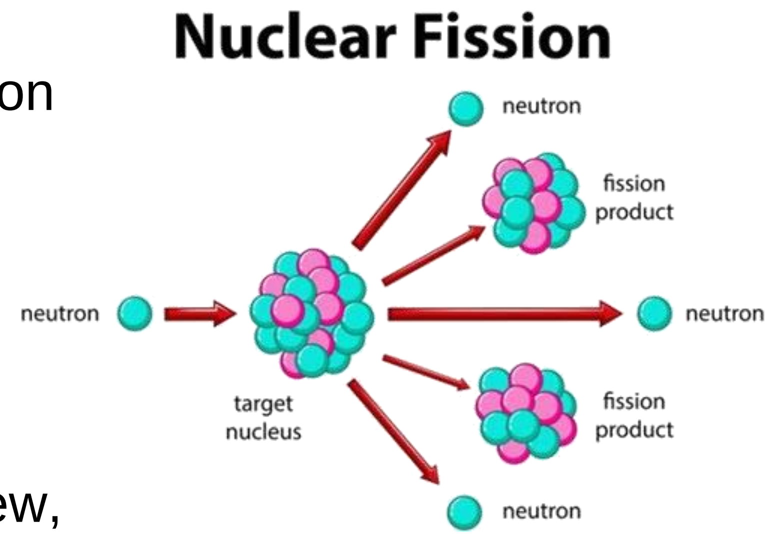
- This class of nuclear reactions becomes significant above 8 MeV [Was]
- It means nuclear reactions of the type ${}^{A}X$ (n,2n) ${}^{A-1}X$

(n,γ) reactions

- The (n,γ) reactions are important at thermal neutron energies, e.g. in nuclear reactors for ^{235}U
- They mean reactions where a nucleus absorbs a neutron, but the process releases a γ photon, $^{A}X(n, \gamma)\, ^{A+1}X$
- This is significant for damage production because the nucleus gets a recoil energy from the emitted gamma particle (since also photons have a momentum $p = E/c$)
- The recoil energy the atom receives is of the order of a few 100 eV's, which is enough for damage production!

Nuclear fission

- Neutrons can also induce nuclear fission
  - This is of course the basis of fission reactor operations
  - Fission can also occur by natural radioactive decay
- From a damage production point of view, it is important to know that the fission products have energies in the 100’s of MeV range
  - This makes them produce damage as “fission tracks” similar to swift heavy ions – more on that in section 2.8.



[Figure from: http://www.iccf11.org/fission-reactor/]

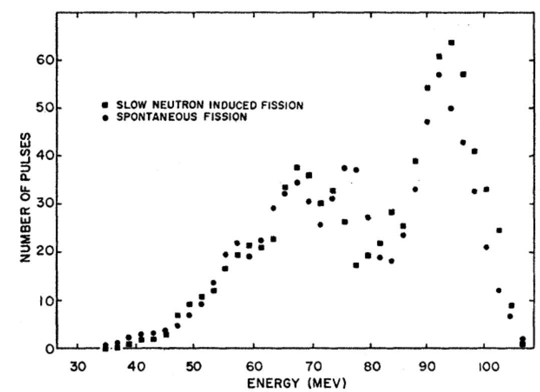
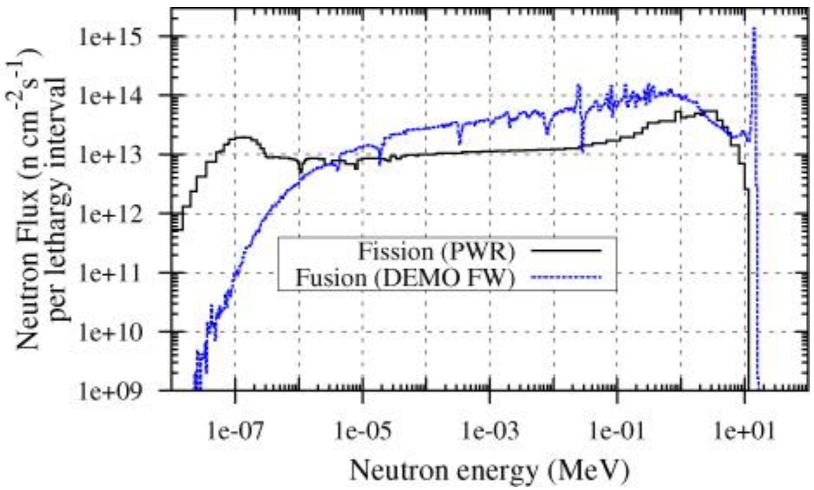


Fig. 1. Histograms of the ionization produced by spontaneous fission pulses of Pu^{240} and by slow-neutron fission pulses of Pu^{239}.

[Segre and Weigand, Phys. Rev. 94 (1954) 157]

Total neutron effects

- The calculation of neutron effects of materials is complicated by the fact that neutron spectra in nuclear reactors are very complicated in shape



[M.R. Gilbert *et al* 2012 *Nucl. Fusion* **52** 083019]

- Different energies have different reactions, so the total damage estimation requires integration over the neutron spectra, all reactions and damage production
  - Doing this is not possible without special software that has all the reactions in a database

2.6 Ions

- Energetic ions interact with matter with 3 different mechanisms:
  1. Collisions with nuclei/atom cores
  2. Collisions with electrons, via several different mechanisms
  3. At high energies via nuclear reactions
- The ion-nucleus collisions are at high energies classical binary collisions under a Coulomb repulsive potential
- Since the ion mass is much higher than that of the electrons, the interaction with electrons slows down the ion, but does not change its path
- The nuclear reactions can (as for neutrons) be very complicated

Ion-atom collisions

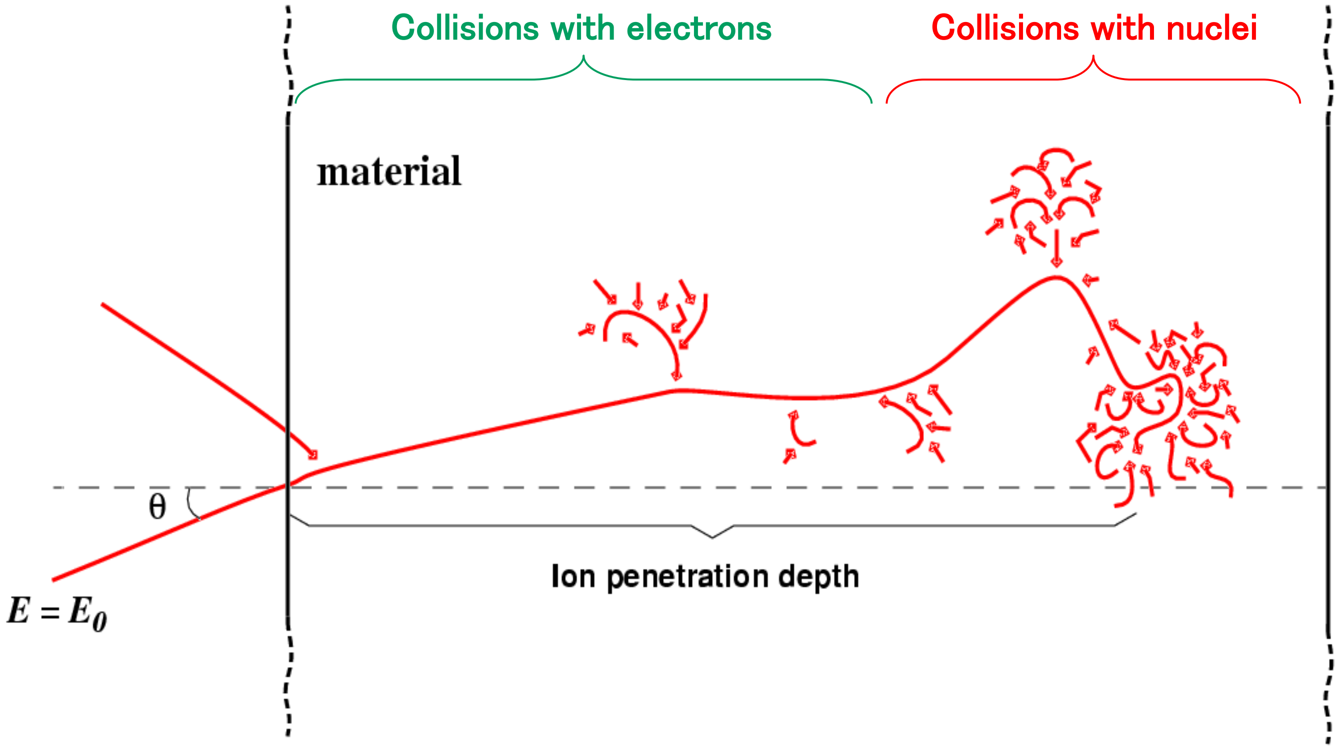
- The same equations as for electrons and neutrons applies also for energy transfer by ions
- The basic classical binary collision equations give the maximum energy transfer (i.e. the energy transfer in a head-on collision) from an ion to an atom of mass m_{atom} as

$$E_{max,atom} = \frac{4\, m_{ion} m_{atom}}{(m_{ion} + m_{atom})^{2}} E_{ion}$$

- The relativistic version of the same equation is only needed for very high ion energies, when the ion velocity approaches the speed of ligh

Picture of slowing down

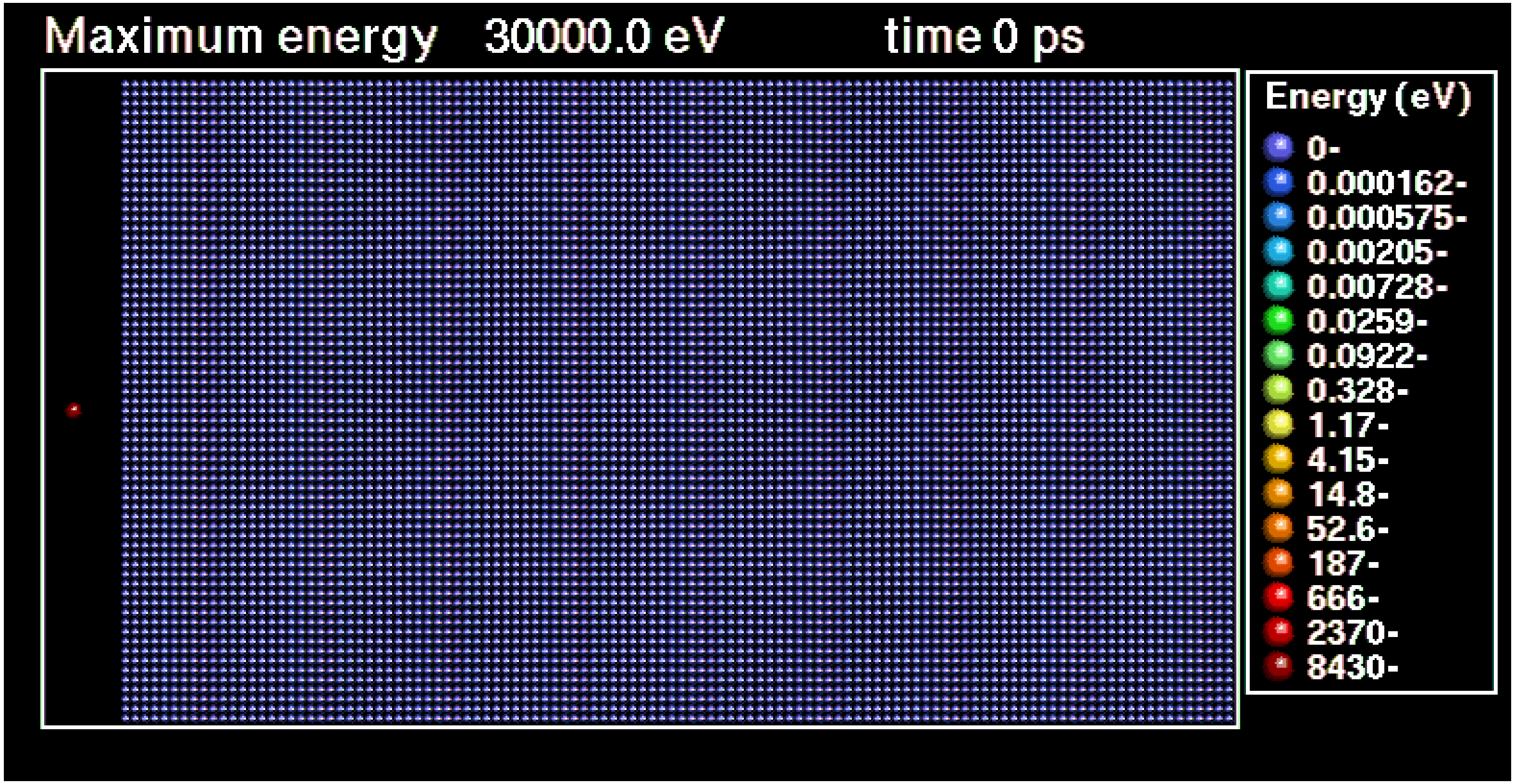
- The slowing down process of an ion impacting on a surface can be roughly illustrated as follows



[Picture: Kai Nordlund]

Animation of slowing down

- Computer simulation for 30 keV Xe impacting on Au
  - Each sphere an atom, cross section picture
  - More on these simulations later



[Animation irradiation.avi]

2.7 Stopping power

- Since the electronic collisions only slow down the ion, and the number of collisions is at high energies large, the effect of the electrons can be considered to be an average frictional force slowing down the ion
- This is known as the **electronic stopping power** (*elektronisk uppbromsning / elektroninen jarruuntuminen*)
- The collisions with ions can also be averaged and then considered a nuclear stopping power (nukleär uppbromsning / ydinten välinen jarruuntuminen)
- The nuclear reactions can also be averaged and considered a nuclear reaction stopping power (*kärnreaktions-uppbromsning / ydinreaktiojarruuntuminen*)

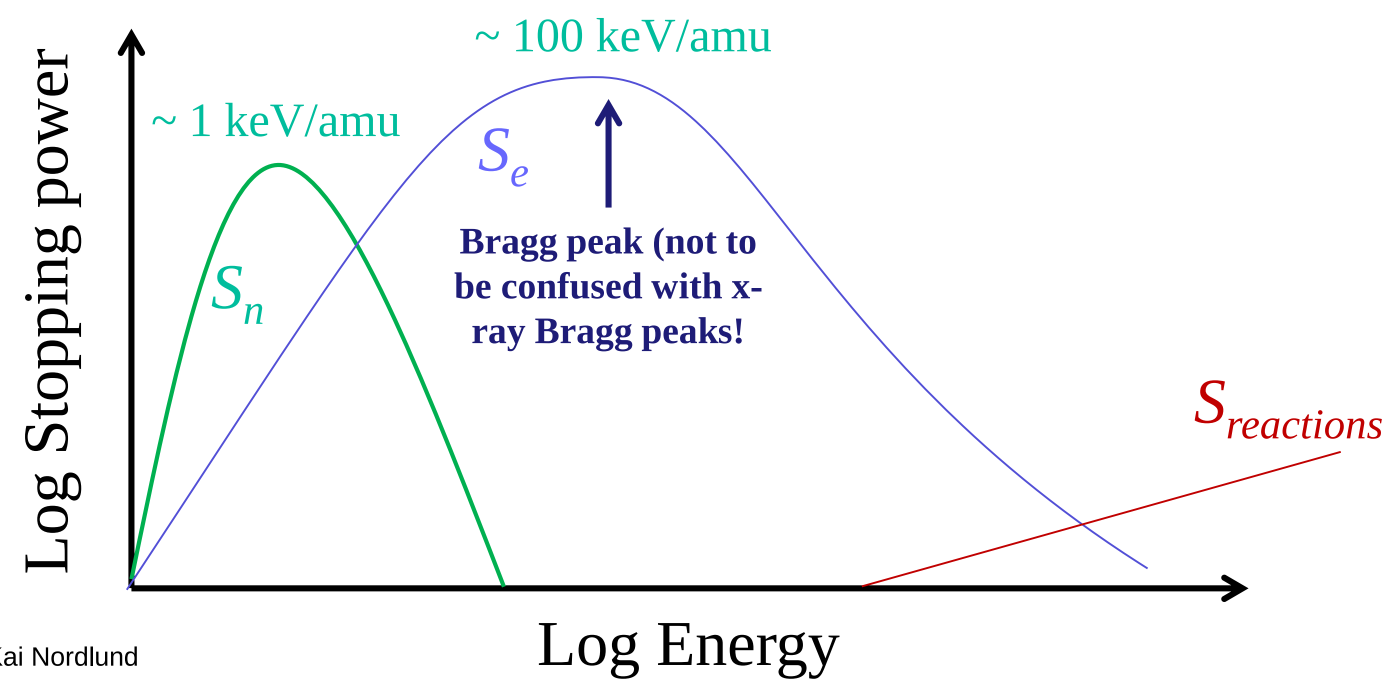
[Key historical reference: [LSS] = J. Lindhard, M. Scharff, and H. E. Schiott, Range concepts and heavy ion ranges, Kgl. Danske Vid. Selskab. Mat. Fyd. Medd. 33 (1963) 1; available online by googling]

Stopping power

- The total stopping power can be then written as

$$S = \frac{dE}{dx} = S_n + S_e + S_{reactions}$$

- The typical energy dependence of these is qualitatively typically as follows:
  - The estimates for the energies are very rough!



Stopping power: terminology notes

- Especially in detector and space physics, a different terminology is used for the same quantities
- **“Linear Energy Transfer (LET)”** is used for total or electronic stopping power
  - Since in this field the interest is mostly in MeV and GeV charged particles, the two are essentially the same
- **“Non-ionizing energy loss (NIEL)”** = Nuclear Stopping Power

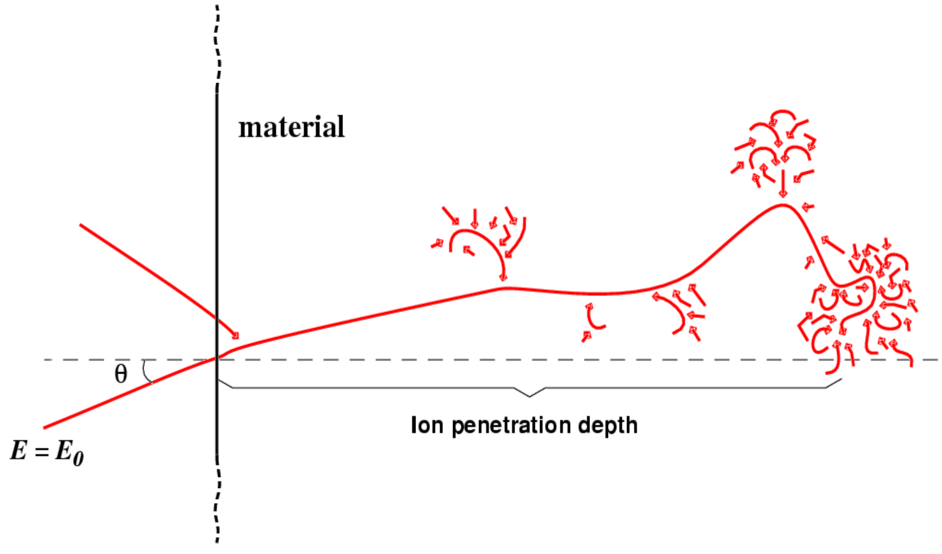
[S. M. Seltzer et al, ”Fundamental Quantities and Units for Ionizing Radiation (Revised)", Journal of the International Commision of Radiation Units and Measurements 11 (2011) 1; ICRU Report 85a]

Stopping power: terminology notes 2

- The historically and still most used term is **“stopping power”**
- One of the leading scientists in the field, Peter Sigmund, argued in the late 1990’s that the word “power” is misleading since the stopping power has units of energy/length, i.e. units of force.
  - Since then, several groups have switched to using the term **“stopping force”** to denote the quantity (without otherwise changing the meaning)
  - However, the still dominant term is still “stopping power”: a Web of Science search for use of the exact terms for 2016-2020 gave 17 hits for “stopping force” but 901 for “stopping power”
- However, one can argue that the term “stopping force” makes sense from the ion point of view, but from a material point of view the standard term makes sense as the “power of the material to stop ions”
  - This usage is also consistent with the use of “stopping power” to description of how armor and bullet-proof vests can stop projectiles shot of them
- In this course, we will stick to using the term **“stopping power”**

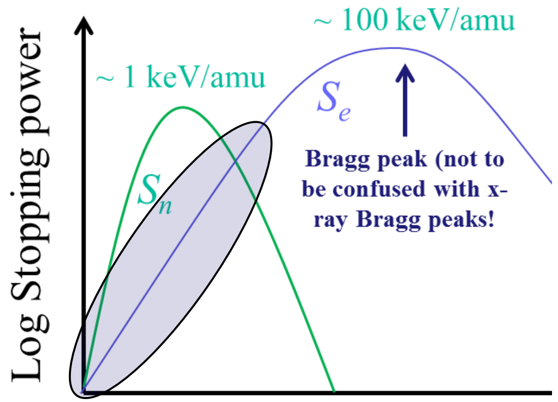
Energy dependence

- The shape of these curves explains why the earlier plot showed straight path atom motion initially, when electronic stopping dominates, and then more curved and dense collisional region when nuclear collisions become significant
  - More on this later



Charge states, ion vs. atom vs. recoil

- An interesting question is, what is the charge state of an ion after it enters a material!?
- At energies corresponding to low electronic stopping powers, below the Bragg peak, the ion loses its initial charge state after penetrating a couple of atom layers, and after that is essentially neutral



  - A more scientific way of putting the same thing: when its velocity is below the Fermi velocity of the material, it moves within the Born-Oppenheimer approximation, and its charge state is the same as if it would be at rest

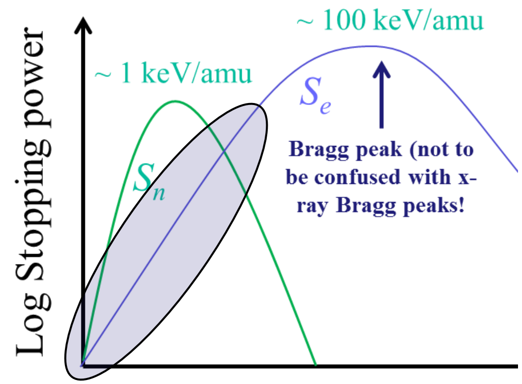
[this is known experimentally from so called hollow atom studies]

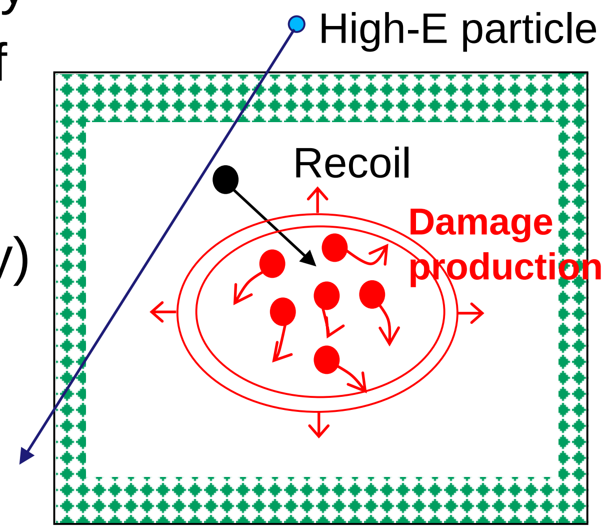
Charge states, ion vs. atom vs. recoil; good news-1

- This implies that for velocities below the Bragg peak, the ‘history’ of the ion/atom does not matter
- Hence damage production by implanted ion, or atomic recoils produced by neutrons, electrons or gammas are only a function of the initial kinetic energy of the moving ion/atom, i.e.

Damage = Damage(Initial Energy)

- Moreover, it means that wrt. damage production below the Bragg peak, “ion=atom=recoil”
- This is a major simplification for predicting damage production!



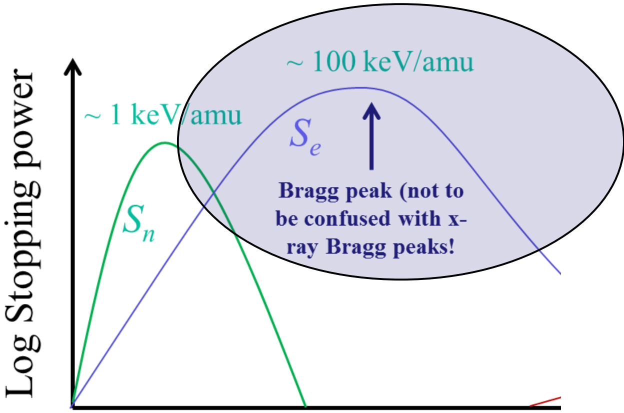


What produces the actual damage? Good news-2

- In metals, excited electrons do not (with very few exceptions) produce damage
- In ionic and organic materials they may, in semiconductors occasionally
- Hence in metals and usually in semiconductors**, all damage is produced by the atomic recoils** – regardless of whether they come from α, β, γ particles or neutrons
  - And also in ionic materials, the atomic recoils often dominate

- Because of this, the separate chapter 5 of this course is dedicated to the issue of damage production by atomic recoils in materials

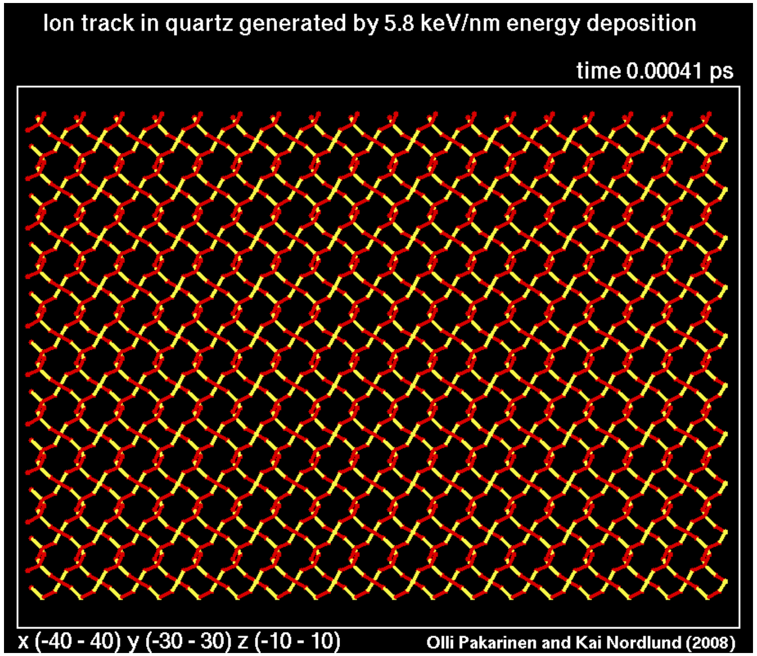
What happens around and above the Bragg peak?



- Around the Bragg peak, the ion has some ‘memory’ of its previous charge state, and above it strong memory
  - The ion is in some highly charged state
- In non-metallic materials, the high level of electronic excitations can produce damage directly in the material
  - Nuclear stopping insignificant = no atomic collisions
- Since the ion at the same time moves in an essentially straight path, this implies that one produces straight tracks of damage in the material

2.8 Swift heavy ions (SHI)

- This regime is known as the **swift heavy ion** (*rask tunga jon / ripeä raskas-ioni**) one
  - Damage known as ion tracks (*jonspår / ioniraita*)*
- Occurs in nature due to natural radioactive decay
- SHI's are produced by high-energy accelerators, now a hot topic for research
  - Fundamental mechanism by which swift ion electron excitations translate into the track damage not known!
    - Intense current research

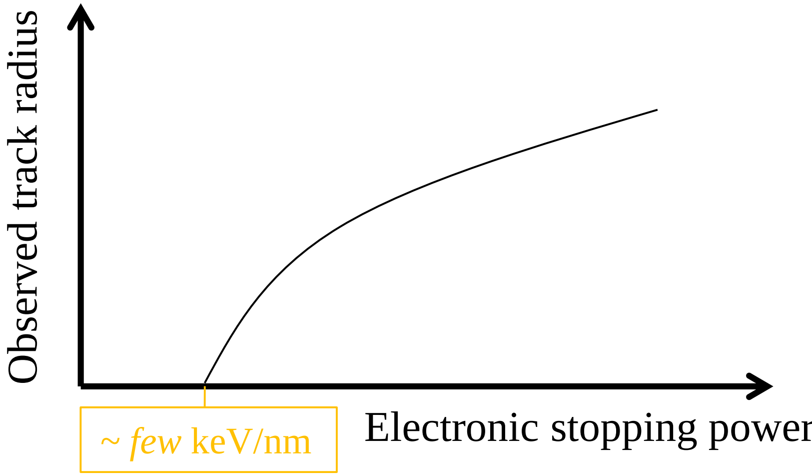


[Animation quartztrack2.avi]

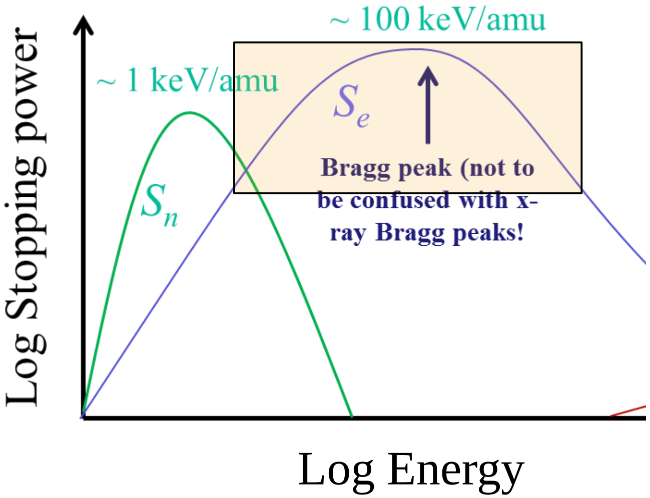
http://www.detectingdesign.com/radiometricdating.html

Ion track production threshold

- The tracks are only observed to be produced above some materials-specific threshold in the stopping power



- Hence a limited energy range around the Bragg peak in stopping



[Review on swift heavy ions: D. Kanjijal, Current Science, 80(12):1560 (2001)

2.9. Computer simulation codes

- There is a range of computer software for simulating energetic particle penetration
  - Many kept up by a software database at the OECD Nuclear Energy Agency (NEA) in Paris
- Ions: BCA codes described in next section
- Neutronics:
  - SPECTR for neutron recoil spectra, NJOY, MCNP, etc.
- Photons and electron-gamma cascades:
  - EGS4 – a modular tool that can be implemented in various geometries, PENELOPE
- Wide-purpose radiation transport on the meso- and macroscopic scale:
  - FLUKA, GEANT4 (especially for radiation detectors in particle accelerators and in space)

Historical note: MCNP dates back to 1956! It is almost certainly the oldest computer code still in regular use [1]

[1] Review of computer simulation of radiation effects: K. Nordlund, http://www.acclab.helsinki.fi/~knordlun/pub/Nor18b.pdf, J. Nucl. Mater. 520, 273 (2019)

What should you have learned from this section?

- You know the basic concepts for radiation penetration, reflection, surface interactions, etc.
- You know how to characterize a particle penetration depth profiles
- You know the basics of how deep different kinds of particles penetrate in materials, and roughly what the physical mechanisms are behind this
- You understand that many different kinds of irradiation eventually produce the final damage by knock-on atoms