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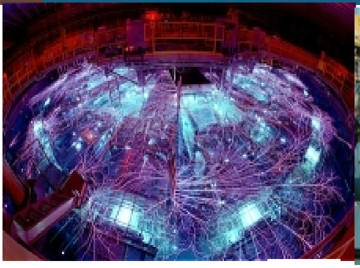
Importance of “Stopping Power” in Assessing Material Damage



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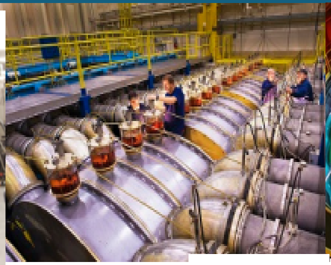

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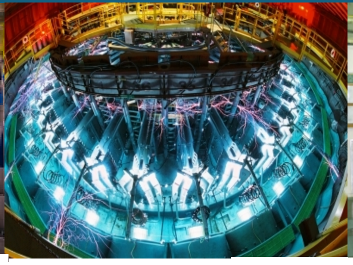

Saturn

ACRR

PRESENTED BY

Patrick Griffin, Laboratory Fellow, Org. 1000

Mar. 3, 2022

Coauthors:

R&A#: xxxx

Approved for Unlimited Release

Name/Org: Patrick Griffin/SNL Date: 02/25/2022
Guidance (if applicable) ______________

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Purpose of Presentation:

- The "stopping power" is a critical ingredient in assessing material damage – but one has to understand the **context** in which it is used. In particular, one must understand the application. This, then, implies a need for details on the: a) application; b) source term, e.g., the ion type and its energy; c) damage metric of concern.
- This presentation defines some **terminology** and establishes an application-focused **framework** for understanding the nuclear data needs in this area.
- We must consider the **uncertainties** in the overall application of interest before we try to prioritize future activities (modeling and experimental) that address our nuclear data needs.

Why this is important?

It helps provide a cost/benefit basis for setting our priorities for better modeling and for gathering more experimental data.

Terminology: Environment / Effect / Damage Mode

Environments

- **Particle type**
  - Neutron (n)
  - Photon (γ)
  - Electron (e)
  - Proton (p)
  - Light Ion (A≤4)
  - Heavy Ion (A>4)
- **Fluence / Intensity**
- **Energy Spectrum**
- **Temporal waveform**
  - Peak rate

Effects

- Heat (Kerma/ Dose/Phonons)
- Ionization
  - Charge
  - Electron creation rate
  - Electron/hole pair creation
- Displacement
  - Frenkel pair creation
  - Defect production
  - Transistor gain
- Transmutation products
  - Activation
  - Impurities
- Physics-based property changes
  - Carrier recombination lifetime
  - Oxide/interface trapped charge

Damage Modes

- Transistor gain degradation
- Photocurrent burn-out
- Threshold voltage offset shift
- nSEE, e.g., upset
- Noise from increased thermal generation of carriers
- CCD charge leakage
- Noise from dark current in sensors/optoelectronics
- Signal attenuation from fiber darkening
- Circuit behavior changes, e.g., from:
  - Resistance increase
  - Capacitance change

- An “**environment**” characterizes the requirements; but “**damage mode**” is surveyed during an assessment.

Definition: Stopping Power

- ICRU 60 defines the "**mass stopping power**" as "the quotient of dE by ρdl, where dE is the energy lost by a charged particle in traversing a distance dl in the material of density ρ."

  SI units of Jm^2/kg but, in radiation damage, is typically reported in units of MeV-cm^2/mg
- It can be partitioned into three components:
  - **Electronic** (or collisional): due to collisions with electrons ; goes into ionization
  - **Nuclear**: due to Coulomb collisions with atoms in a material; goes into breaking interatomic bonds or generating lattice phonons
    - The term "nuclear" here **does not** refer to the strong or weak nuclear force.
    - Sometimes separated into elastic and inelastic Coulomb energy losses due to the ion interacting with the nucleus of the lattice atoms.
  - **Radiative**: emission of bremsstrahlung in the presence of electric fields of nuclei and electrons
- "**linear stopping power**" is the product of the mass stopping power and the material density. SI units of J/m, typically reported in keV/µm.

While stopping power refers to the energy lost, the linear energy transfer (LET) refers to the energy absorbed. The unrestricted LET is the electronic stopping power.

Examples of Material Damage Applications

Application Area

- **Spacecraft damage**
  - Solar panels
  - Internal electronics
  - Sensors
- **Outer planet exploration**
  - Trapped radiation belts
- **Nuclear Propulsion**
  - Material embrittlement
  - Control electronics
  - Passengers
- **Commercial fission reactors**
  - Material embrittlement
  - Control electronics
  - Personnel

Material of Interest

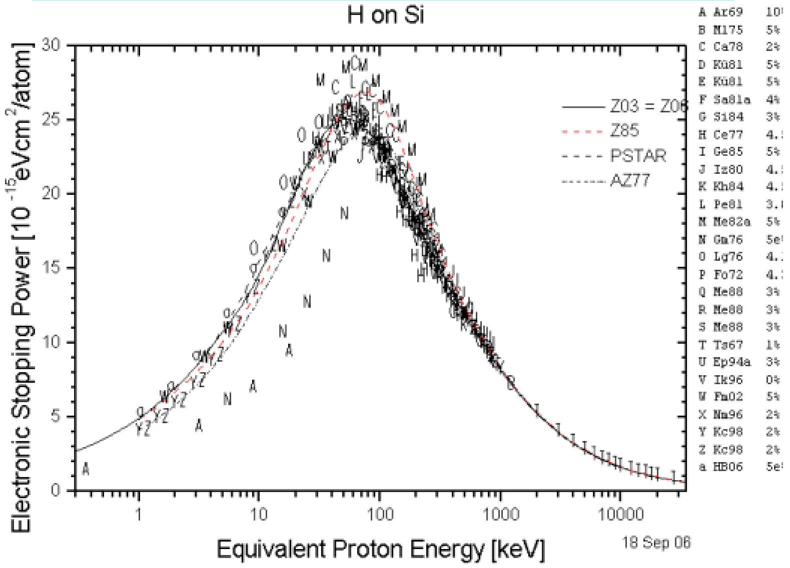
- Personnel Safety (**tissue**)
  - ISS space station
  - Trip to Mars
  - Aircraft crew
- Electronics (**Si, SiO_2, GaAs**)
  - Upset in mission-critical electronics, i.e., aircraft controls
  - Ground computers, i.e., SRAM in supercomputers or ND monitoring
- Sensors - DSP, SBIRS, CHAMP, LANDSAT, GOES (**HgCdTe, InP, $LiTaO_3$, InSb, Si, Ag**)
  - Charge-coupled device (CCD)
  - Focal plan arrays (FPA)
  - Mirrors

Take-away: The application determines the relevant radiation source type/energy; the material; the damage mode of interest.

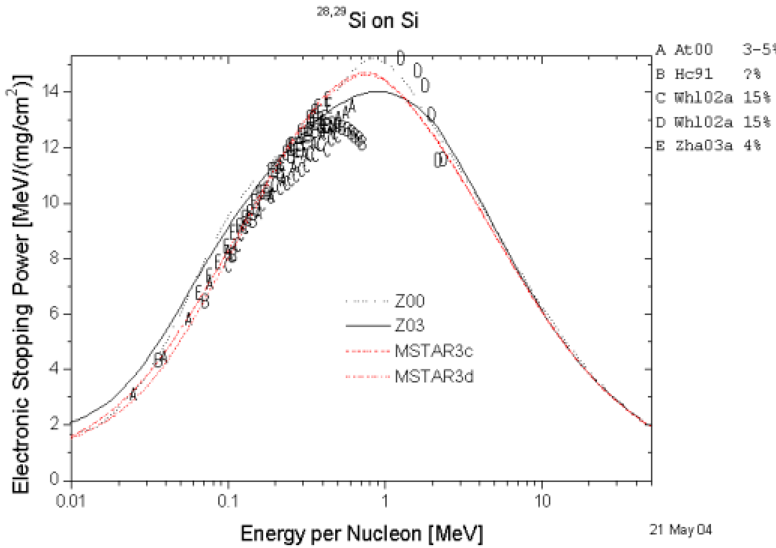
Stopping Power

- Stopping power shows a peak wrt energy – the Bragg peak.
- Database supported at IAEA: . https://www-nds.iaea.org/stopping/
  - See following presentation by Claudia Montanari

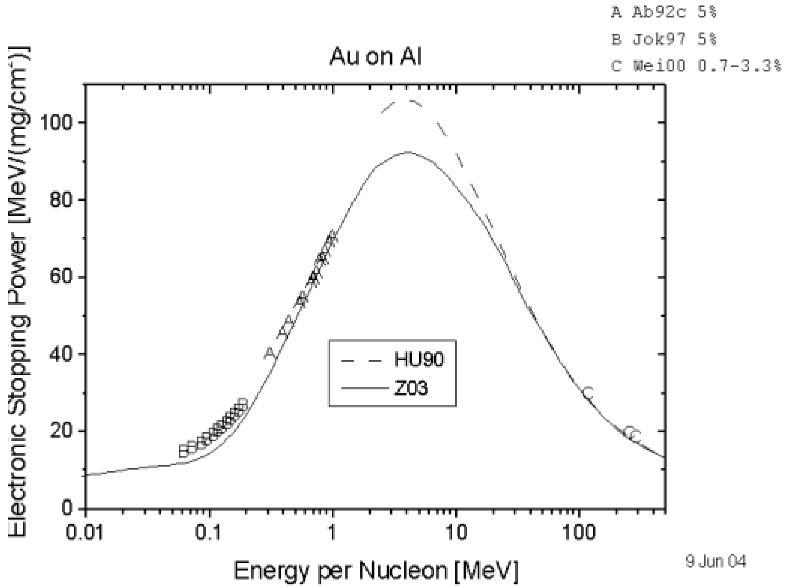
Proton in Silicon
- **units = 1E-15 eV-cm²/atom = 0.02144 MeV-cm²/mg**
- **peak = ~ 100 keV ~ 0.536 MeV-cm²/mg**



Si Ion in Silicon
- **units = MeV-cm²/mg**
- **peak = ~ 1-MeV/nucleon ~ 14 MeV-cm²/mg**



Au Ion in Aluminum
- **units = MeV-cm²/mg**
- **peak = ~ 5-MeV/nucleon ~ 100 MeV-cm²/mg**

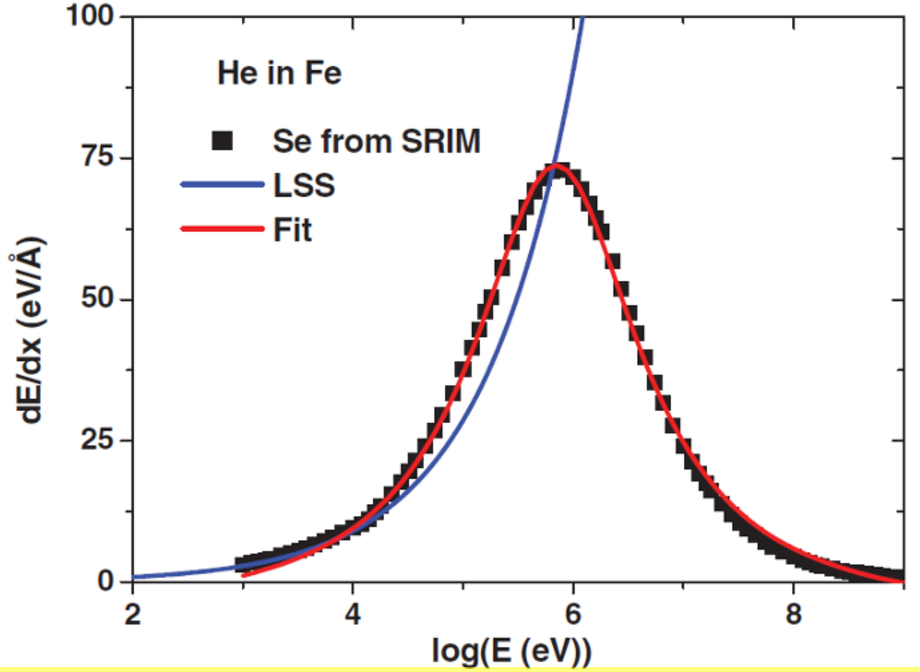


Scaling rules exist when x-axis displayed as energy/nucleon.

Calculation of Stopping Power

- Work in 1963 by Lindhard, Scharff, and Schiott (LSS).
  - Based on Thomas-Fermi screening function using local and non-local electronic energy loss.
  - Breaks down when you impact more than the Bohr velocity to the lattice recoils; LSS limitation $E_{ion} < 24.8 \bullet Z^{4/3} \bullet A$ (keV).
- The Zielger, Biersack, and Littmark (ZBL) semi-empirical approach is used in codes such as SRIM.
- Other codes:
  - MSTAR, H. Paul and A. Schinner, 2003
  - DPASS, Sigmund and Schinner, 2020
  - CasP, Grande and Schiwietz, 2012

Differences between physics-based models and empirical fits is an area of active research.



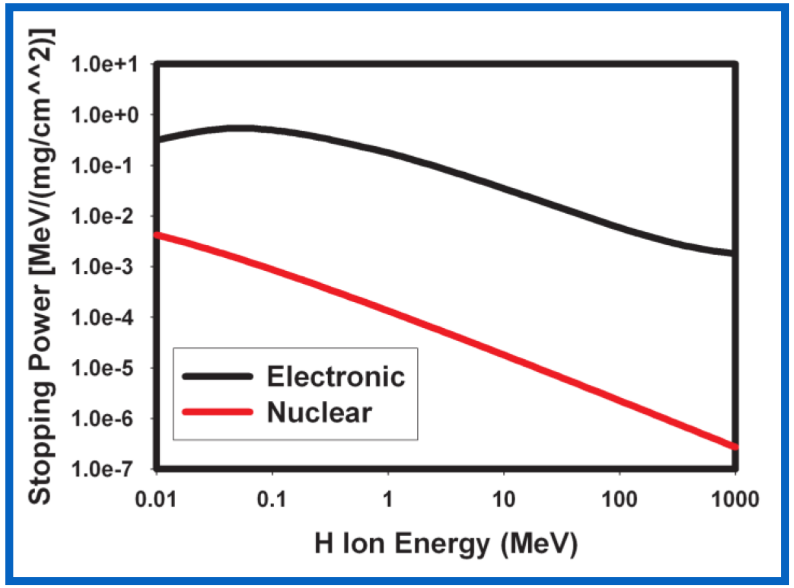
Source: SC. Ortiz, et al., Rad. Eff. And Defects in Solids, Vol. 169, 2014.

Electronic vs. Nuclear Stopping Power

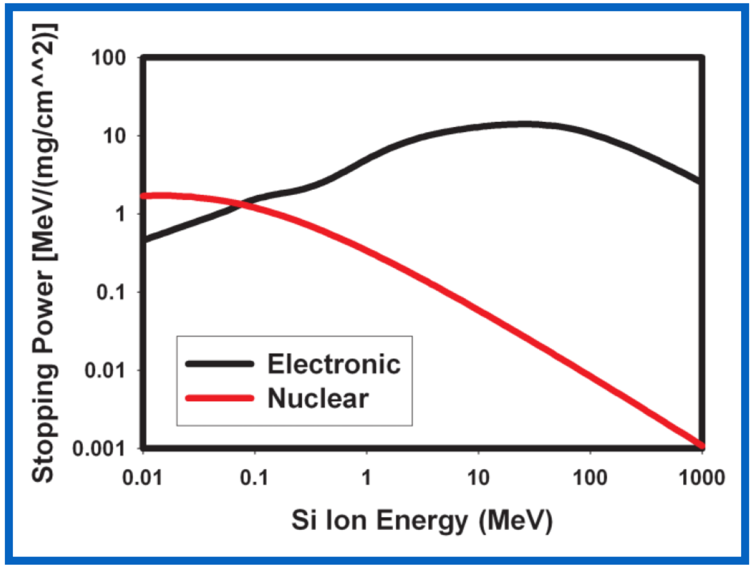
- Damage mechanisms usually depend upon **either** the electronic or the nuclear component of the deposited energy.
- Ionization dominates at high recoil energies
- Ionization more important for high-Z ions

- **This is a partition of the differential energy deposition wrt an ion track**

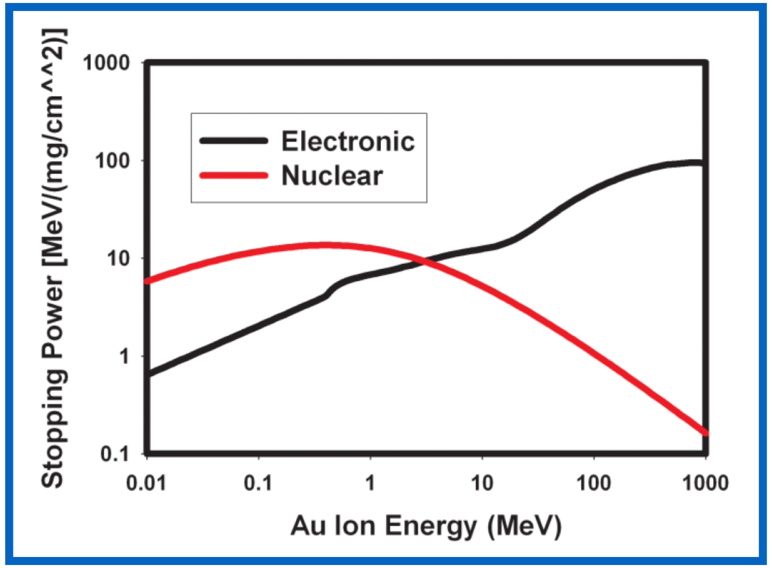
H in Si



Si in Si



Au in Si



Damage Partition Function

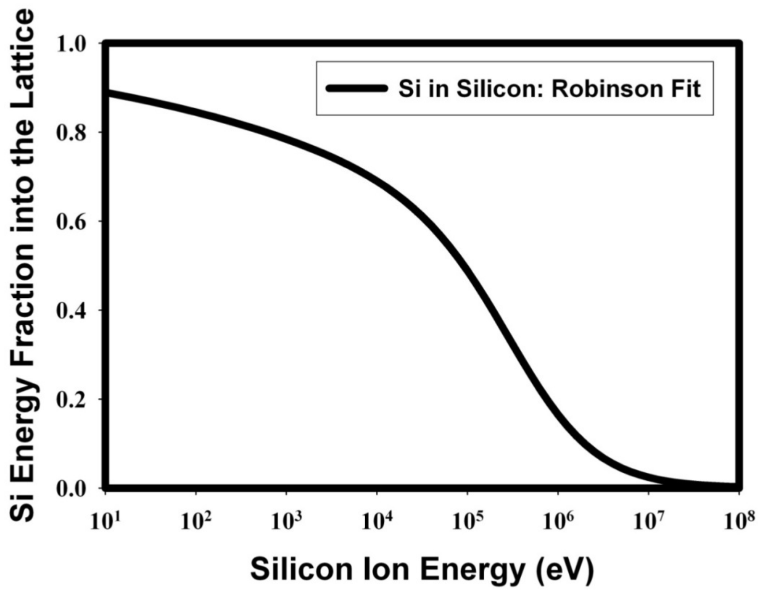
- For the assessment of material damage, we often use a metric based on the energy going into **ionization** and/or **displacement** (into the lattice) when integrated over the path of the ion.
- The fraction of the energy going into the lattice, when integrated over the ion slowing down path, is called the "damage partition function".
- Codes such as NJOY use the Robinson fit, which is based on LSS stopping powers, to determine this.

$$Q = 1/[1 + k_L\, g(\varepsilon)],$$

$$g(\varepsilon) = \varepsilon + 0.40244\, \varepsilon^{3/4} + 3.4008\, \varepsilon^{1/6}.$$

$$k_L = 0.0794 \frac{Z^{2/3} Z_1^{1/2} (A + A_1)^{3/2}}{(Z^{2/3} + Z_1^{2/3})^{3/4} A^{3/2} A_1^{1/2}}.$$

- A: atomic mass
- Z: atomic number
- l: lattice atom



Nuclear Data Needs (1/3):

- **Experimental data for Stopping Power:**
  - Data for more ions on materials on interest, e.g., GaAs, GaN.
  - Many materials/ions are not covered by the IAEA / H. Paul's database. So, we fall back on calculations (physics-based or semi-empirical) with no clearly documented associated uncertainty.
- **Validation data for the partition function:** i.e., the partition of the stopping power, either differential or integrated over the ion path.
  - **Robinson** formalism is not good at high energy. **Akkerman** formalism only fits Si. We need **simple scaling rules** for processing code interface.
  - Need **uncertainty** (std. dev. & energy-dependent correlation matrix) in the stopping power and in the damage partition function.
  - Need **model-based estimates** and **validation data** for this uncertainty – there is a strong energy-dependent correlation which must be characterized for integral damage metrics.

Our nuclear data needs start with DATA! Data must always have an associated uncertainty.

What About the Damage Partition for a Neutron?

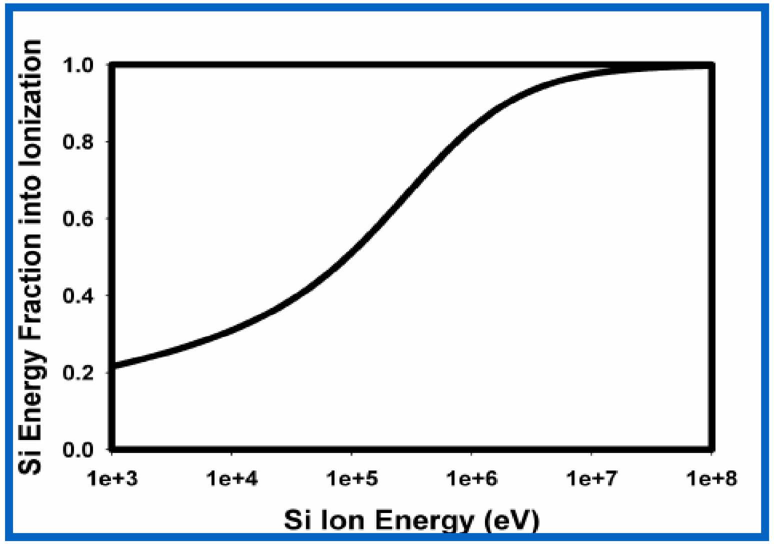
- The nuclear data files, e.g., ENDF/B, provide the probability of interaction.
- The nuclear data files and/or two-body kinematics provide the recoil ion energy.

- Total Dose ≠ Ionizing Dose
- Most applications require a correction factor, e.g., Si PIN response, trapped oxide charge.

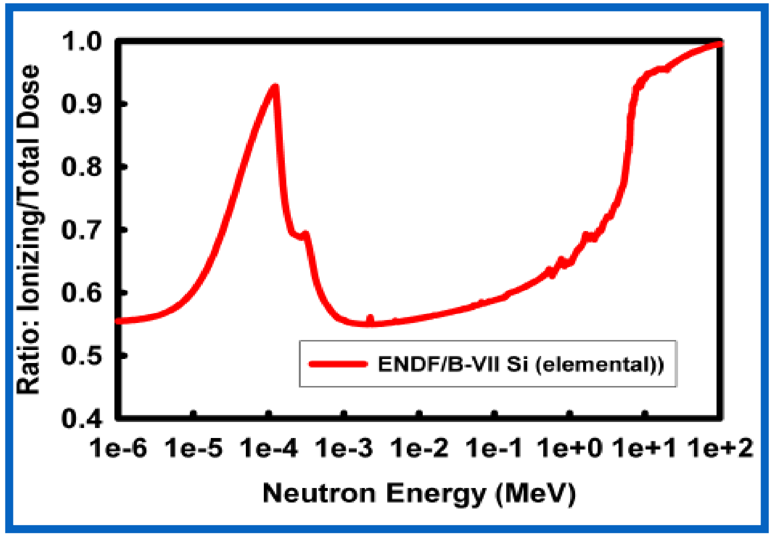
Si Neutron Response

Spectrum	% n-dose	% n- dose Ionizing	% Effect on Si TID Rsp.
SPR-III CC	24.7	56.74	14.0
WSMR FBR 6”	32.9	57.4	18.9
ACRR CC	7.32	55.66	4.07
ACRR Pb-B	26.8	52.95	14.2

SI Ion Ionization/Total Dose

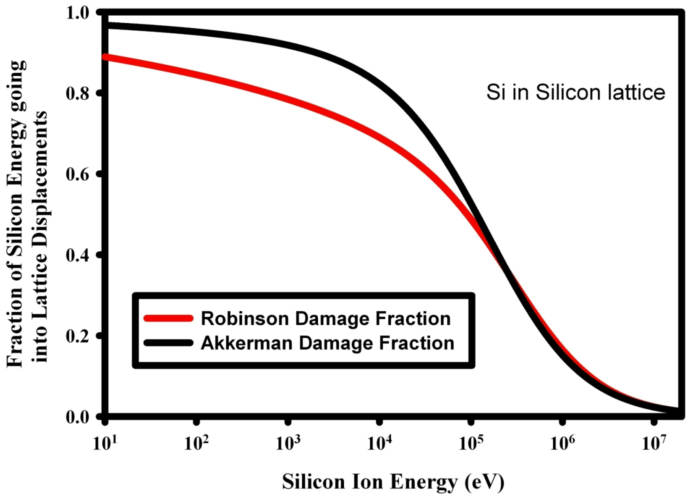


Si Neutron Ionization/Total Dose

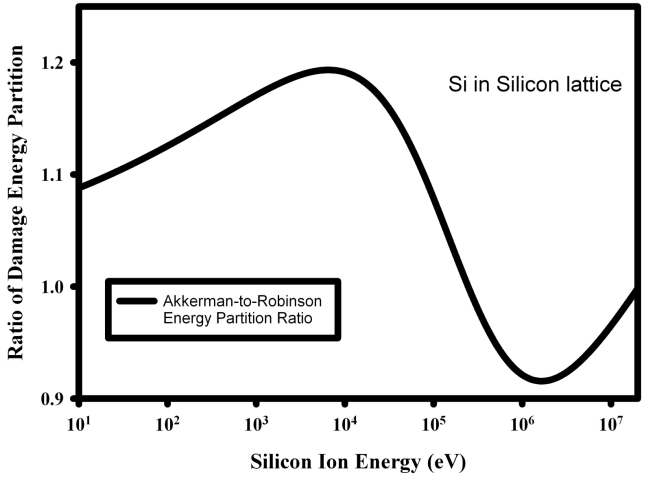


Variation in the Silicon Ion-based Partition Function

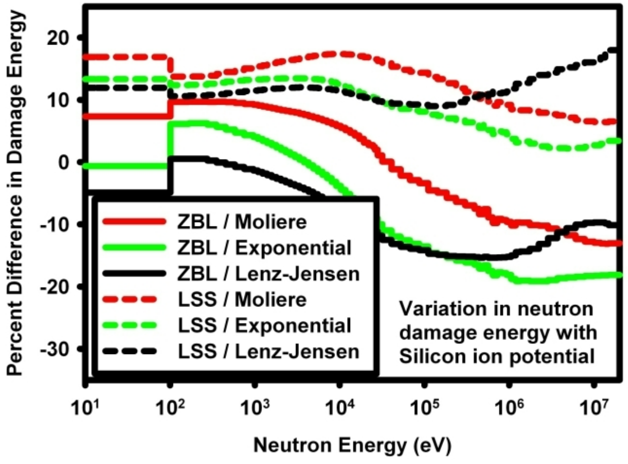
- What do calculations tell us about the variation in the partition function?



Comparison of damage partition function



Ratio of damage partition function



Effect of potential on neutron damage partition function

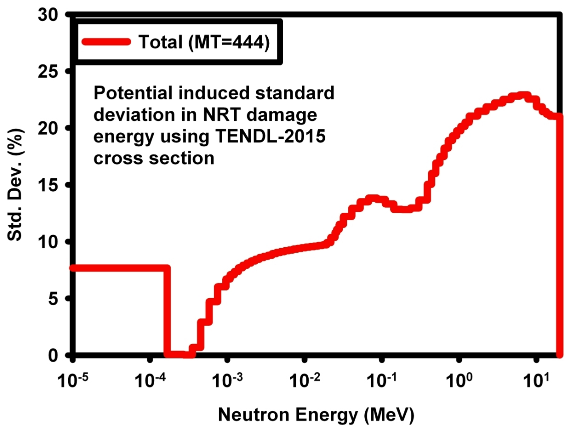
Main difference is LSS (Robinson) vs. ZBL (Akkerman) potential/screening.
Effect of potential changes based on MARLOWE BCA calculations.

For Si, potential variations result in a +/- 20% % variation (std. dev.).

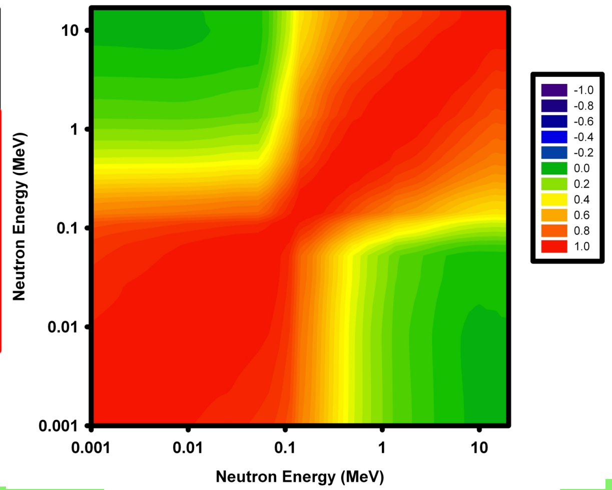
There is a strong energy-dependent correlation in the damage partition function

Neutron-based

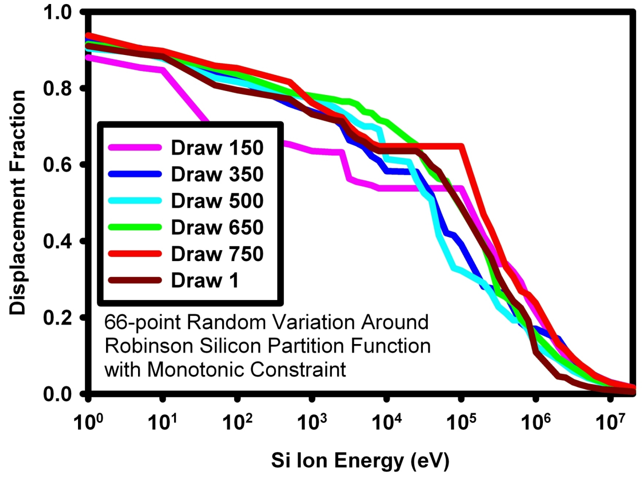
Si ion-based



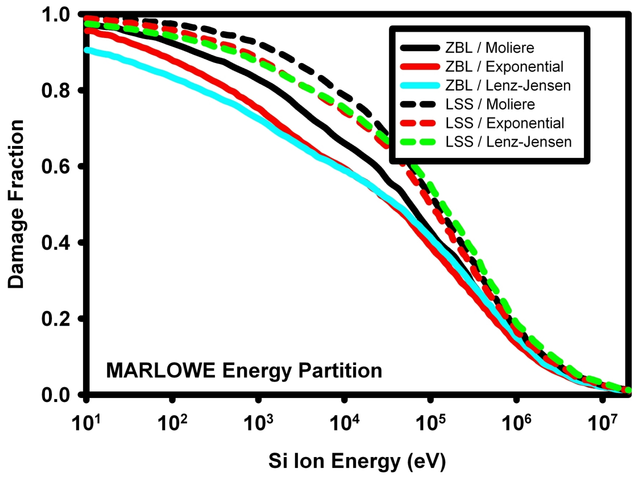
Std. dev. in Si damage partition function



Correlation Matrix



Statistical draw of damage partition function



Effect of potential on damage function

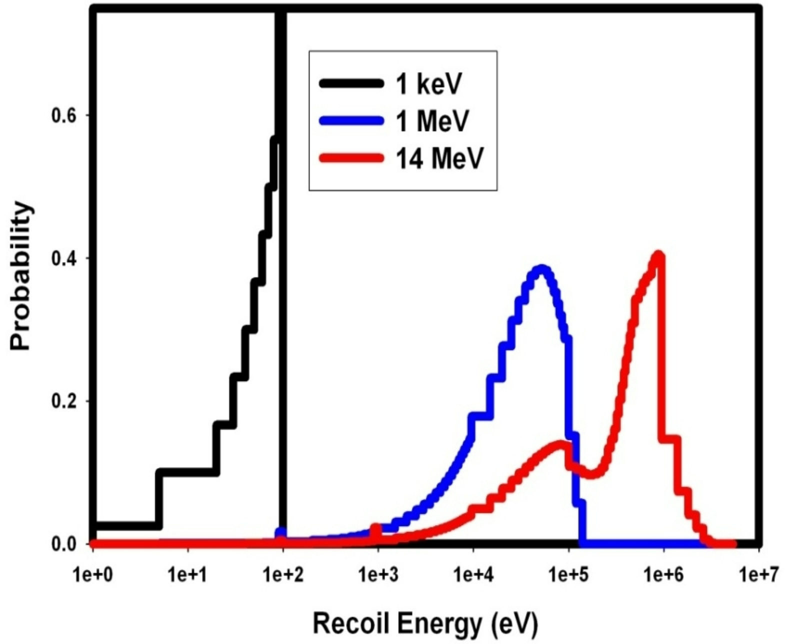
Neutron-based uncertainty based upon TENDL-2015 random ENDF files for recoil spectra.
Si ion-based uncertainty based on MARLOW-based BCA calculations.

A strong energy-dependent correlation can result in a 2X change in integral uncertainties!

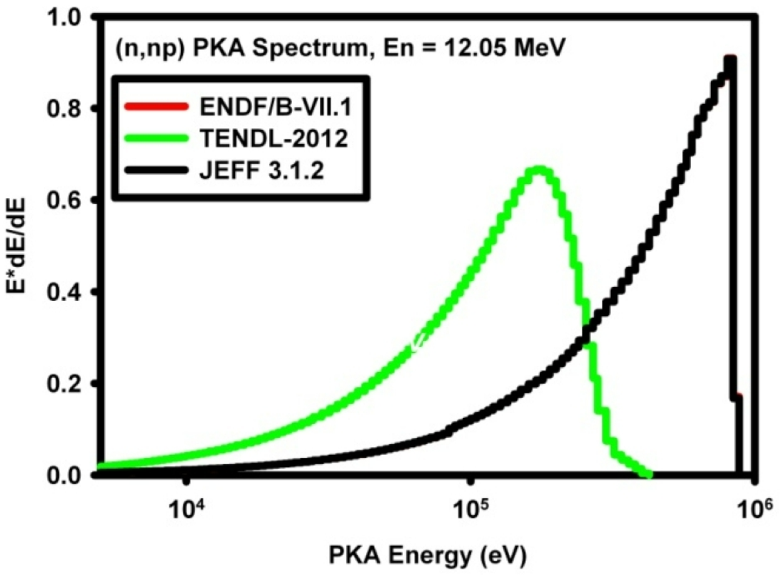
Uncertainty in Recoil Spectra

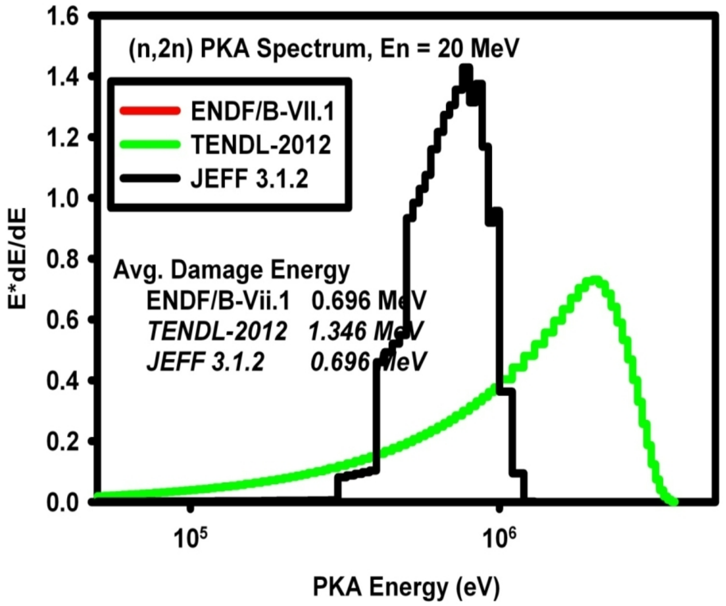
- The recoil spectrum is complex.
- Different reactions has different recoil spectra.
- The recoil spectra for some reactions have a large model-based uncertainty.

Complex PKA Recoils



Large Uncertainty for some reactions



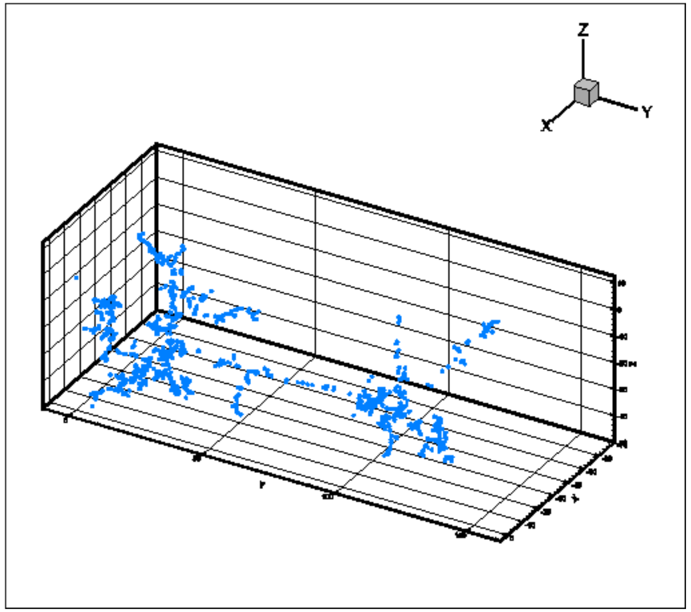


We need good recoil spectra for non-elastic channels with quantified uncertainties.

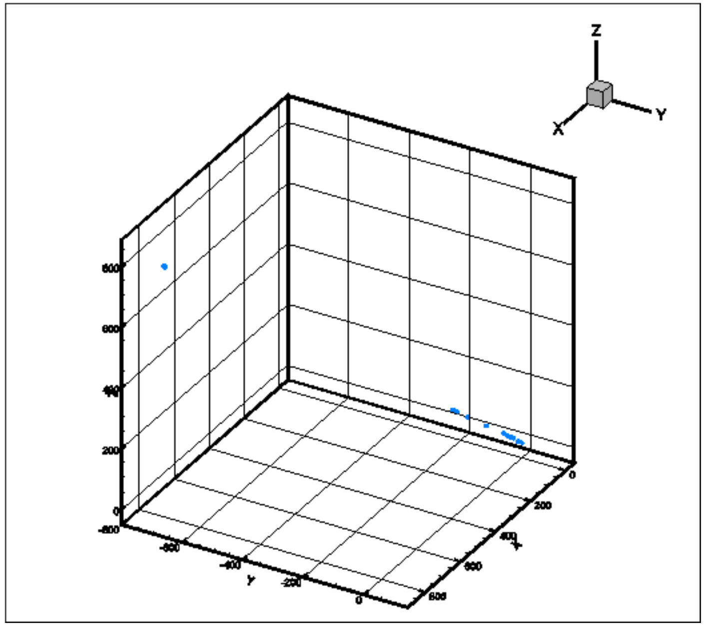
Statistical Process: there is a significant cascade-to-cascade variation in FPs

Sample cascades for a 100 keV Si ion in Si

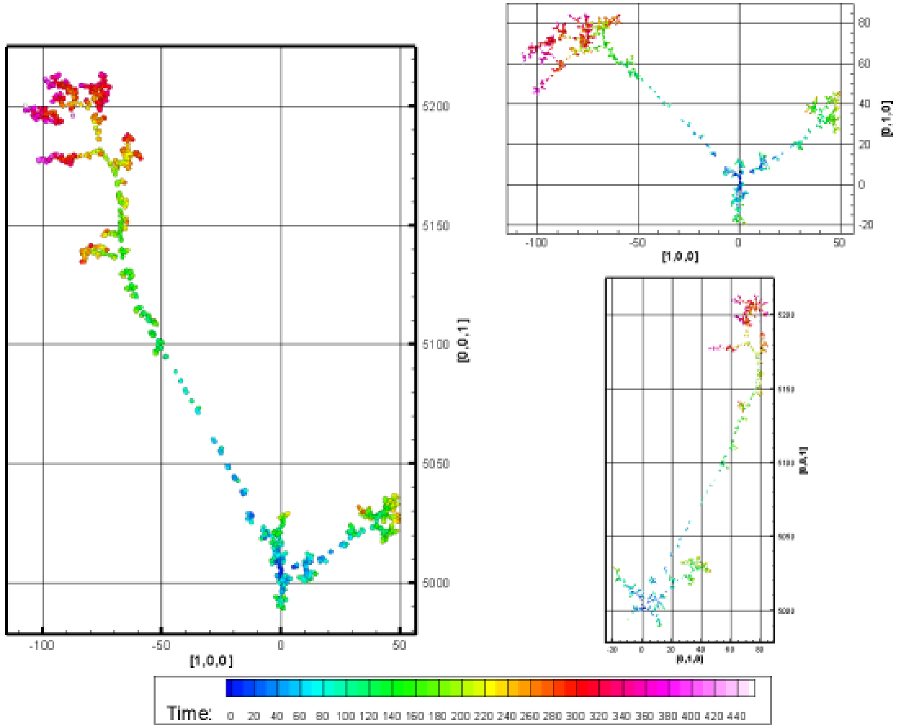
925 distant FP
(Vacancies only)



40 distant FP
(Vacancies only)



Single 100 keV Si ion track



MARLOWE calculation by P. J. Cooper

The variation in damage metrics due just to the cascade process can be significant.

Probability Distribution for FP Production

pdf for Si Ions

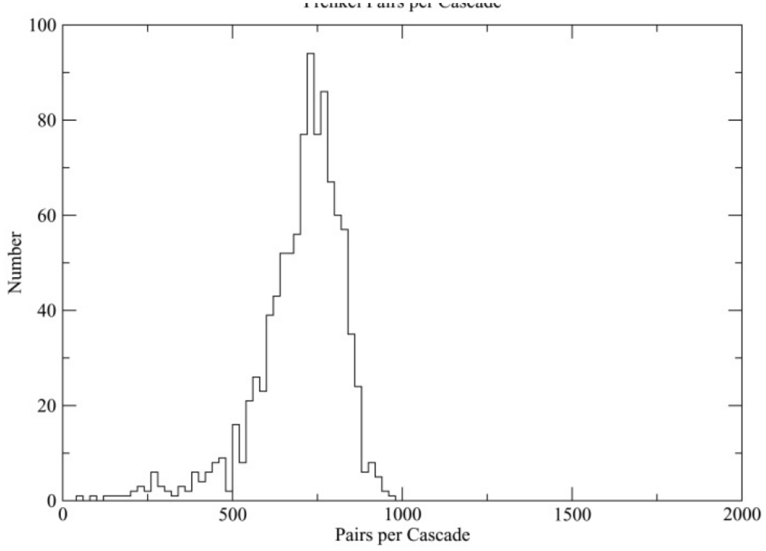
pdf for <u>1-MeV</u> neutrons in Si.

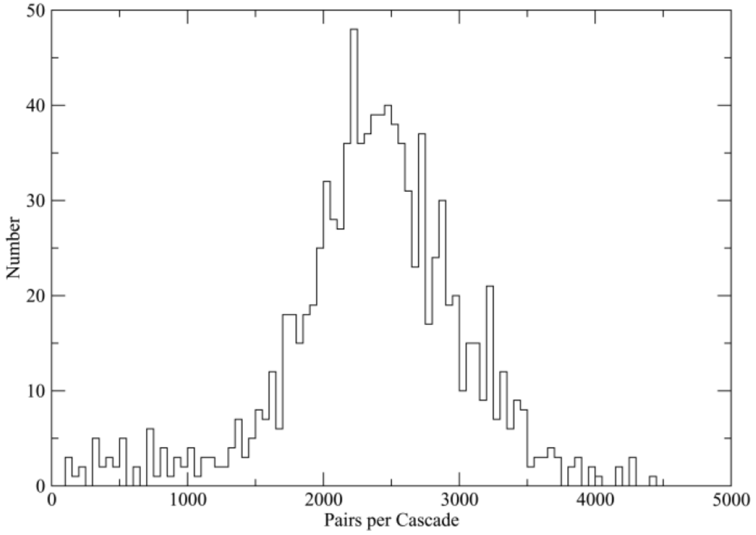
100 keV
<FP> = 680; FWHM = 129

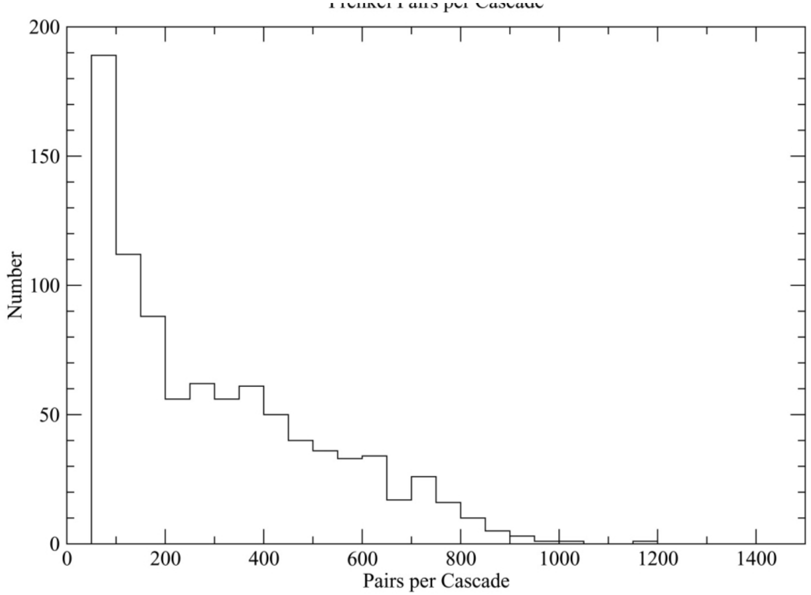
1 MeV
<FP> = 2320; FWHM = 656

<FP> = 244; FWHM = 216

$\langle E_{recoil} \rangle$ = 38 keV; FWHM = 31 keV







The distribution is critical! It is not a normal distribution.
The variation in the pdf can be as large as the mean value.

Nuclear Data Needs (2/3):

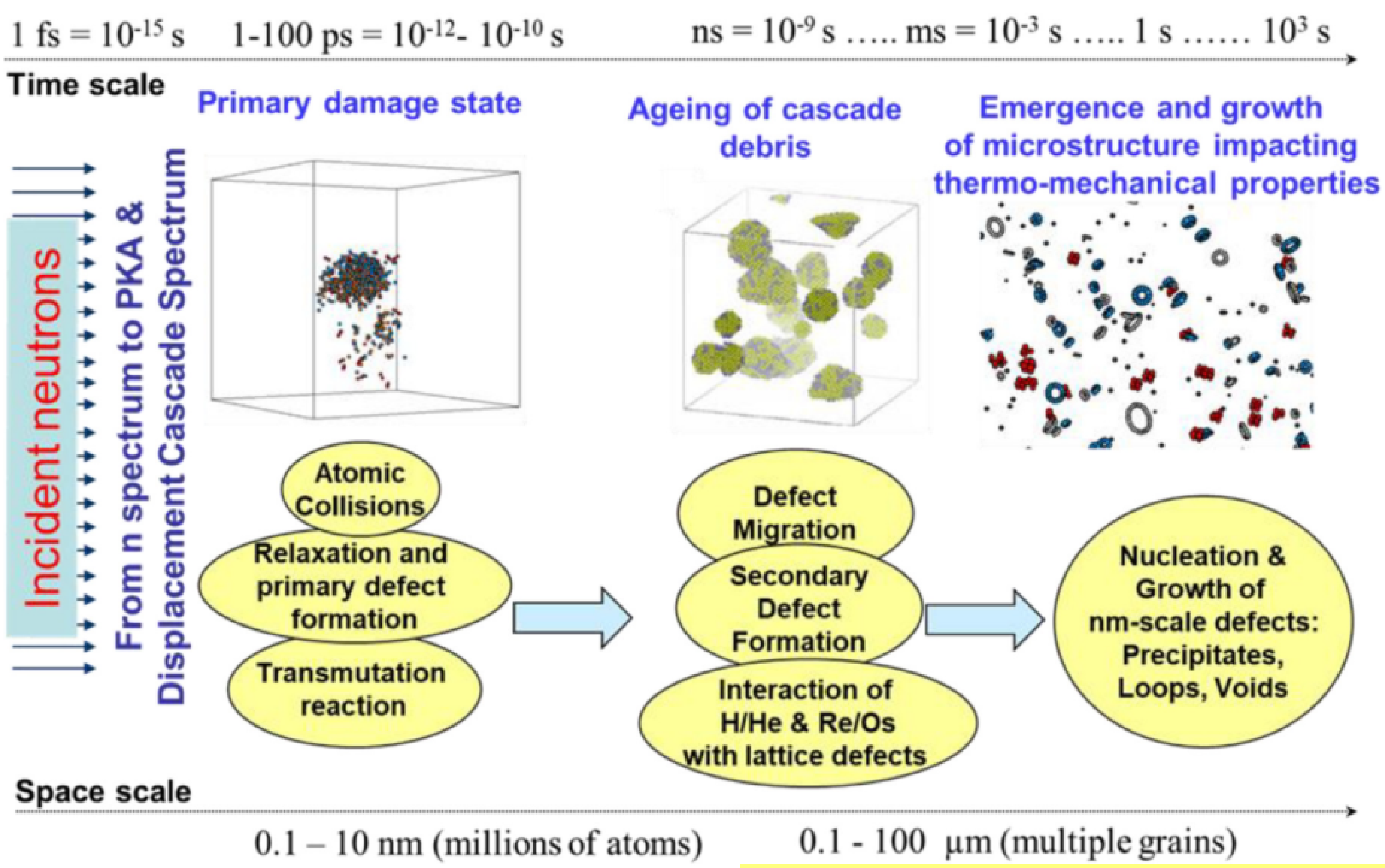
- **High quality neutron recoil spectra** in the ENDF/B MF6 file are the starting point.
  - We need MF6 for all isotopes. Done for calculated TENDL-2021, but we would like to ensure that the evaluator input is based on available data.
  - Need uncertainty in MF6 data. TENDL random draws address some of this – but is believed to miss some model-based uncertainty components.
  - Correlations of cross section between reaction channels can be significant.
- Stopping power is not enough, we also need **details of the initial damage structure** to support:
  - **track structure** modeling and **e/h creation** – as statistical distributions
  - **evolution of defects** and charge state of specific defects

We need a balance in the uncertainty of elements of the input characterization.

Stopping power uncertainty: depends upon the neutron-induced recoil spectra; influences the evolved damage state.

The "Stopping Power" is only the Starting Point for Material Damage

- **Displacement:** Frenkel pair creation
  - Defect migration
  - Defect charge state
  - Nucleation and growth
  - Gas bubble formation and release
- **Ionization:** Charge generation
  - Charge collection
  - Charge recombination
  - Bond breaking
  - Bond interactions



Source: Castin et al., J. Nucl. Matl., Vol. 562, 2022

The physics in the modeling must go beyond the initial/primary event (Frenkel pair creation and ionization) and address defect migration and growth as well as charge collection, transport, and bond breaking.

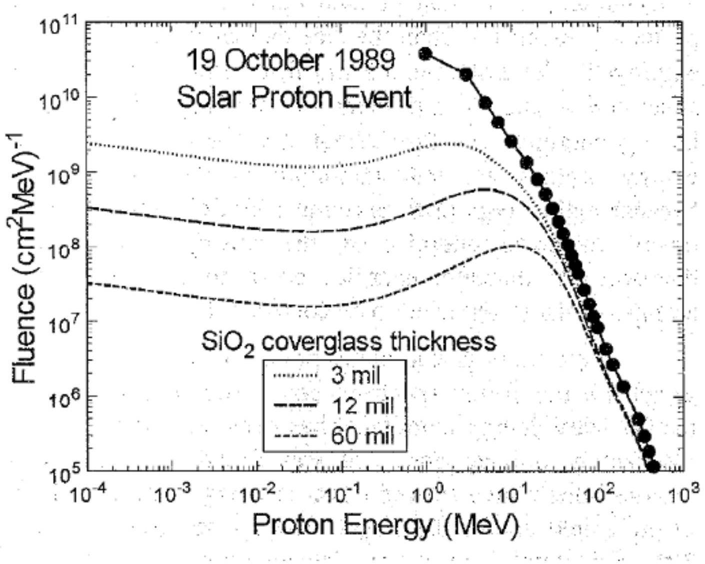
Nuclear Data Needs (3/3):

- Given the stopping power initial conditions, we need **validation data** for advanced modeling of **damage-relevant attributes**.
  - **Defect-specific diagnostics** for radiation damage to materials. We have DLTS for silicon, but lack meaningful defect-specific damage signatures (DLTS, DLOS, NMR, ESR, FT-IR, PAS, PL) for GaAs or GaN.
  - **Validation data** for complex damage modes.
    - Need to model displacement-induced **defects at times later** than MD can address, i.e., mean rate theory and kinetic Monte Carlo. We lack sufficient data to constrain the number of free parameters appearing this theory.
    - The **charge state for defects** can be critical to the damage mode, e.g., recombination lifetime. Yet, current MD modeling does not consider this coupling.
    - **Bubble/void formation** from liberated gases [proton (H) and alpha (He)] can be critical to material embrittlement. Sudden **gas release** can be initiated by a lattice stress. Slow release is also possible.

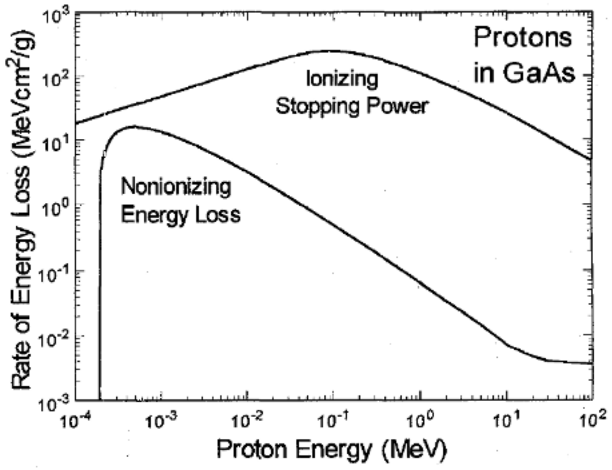
Prioritize nuclear data needs based on the particles/energy and in the context of the application-specific relevant damage mode.

Consider the application focus when you assess the nuclear data needs

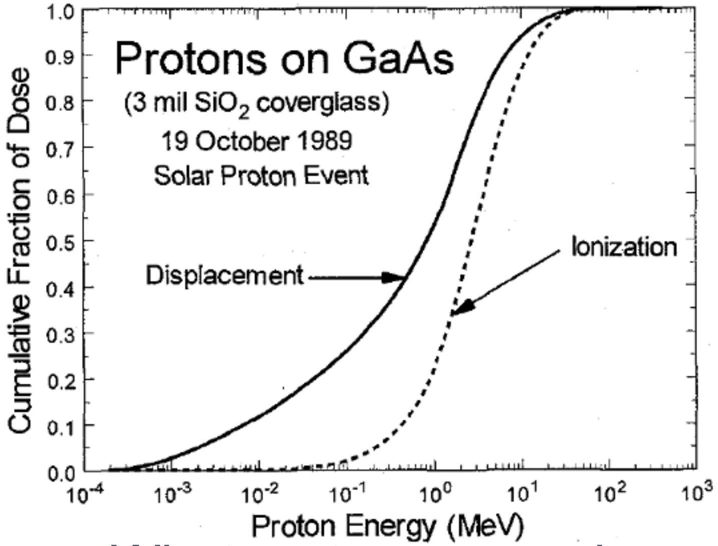
- e.g., solar flare displacement damage in GaAs solar panels



Solar flare protons source term at the test object location, i.e., effect of coverglass



Damage partition for in GaAs for Solar Flare



What proton energy is important?

Source: S. Messenger et al., IEEE TNS, Vol. 44, 1997

Observation: Consider the application-specific cumulative damage. For solar flare damage to spacecraft solar panels, 1 – 10 MeV protons are the primary damage consideration.

Elemetns to Consider in the Analysis Flow

- **Source term**: What is the ion type/energy distribution?
  - Application Source Term:
    - Cosmic-ray
      - Atmospheric-induced neutrons [half-life 10.2 m for “free neutrons”; stable within nucleus]
    - Coronal Mass Ejection
    - Solar protons/electrons
    - Trapped Belts [Earth; Outer planets]
  - Simulation:
    - Ions [DT; proton accelerator; cyclotron-produced ions]
    - Neutrons [fission reactors; spallation neutron source]
- **Energy deposition** – Dynamics of the stopping power
- **Damage evolution** – Metrics of interest

Prioritize the nuclear data in the application of the stopping power in materials within the context of the mission.

Questions?

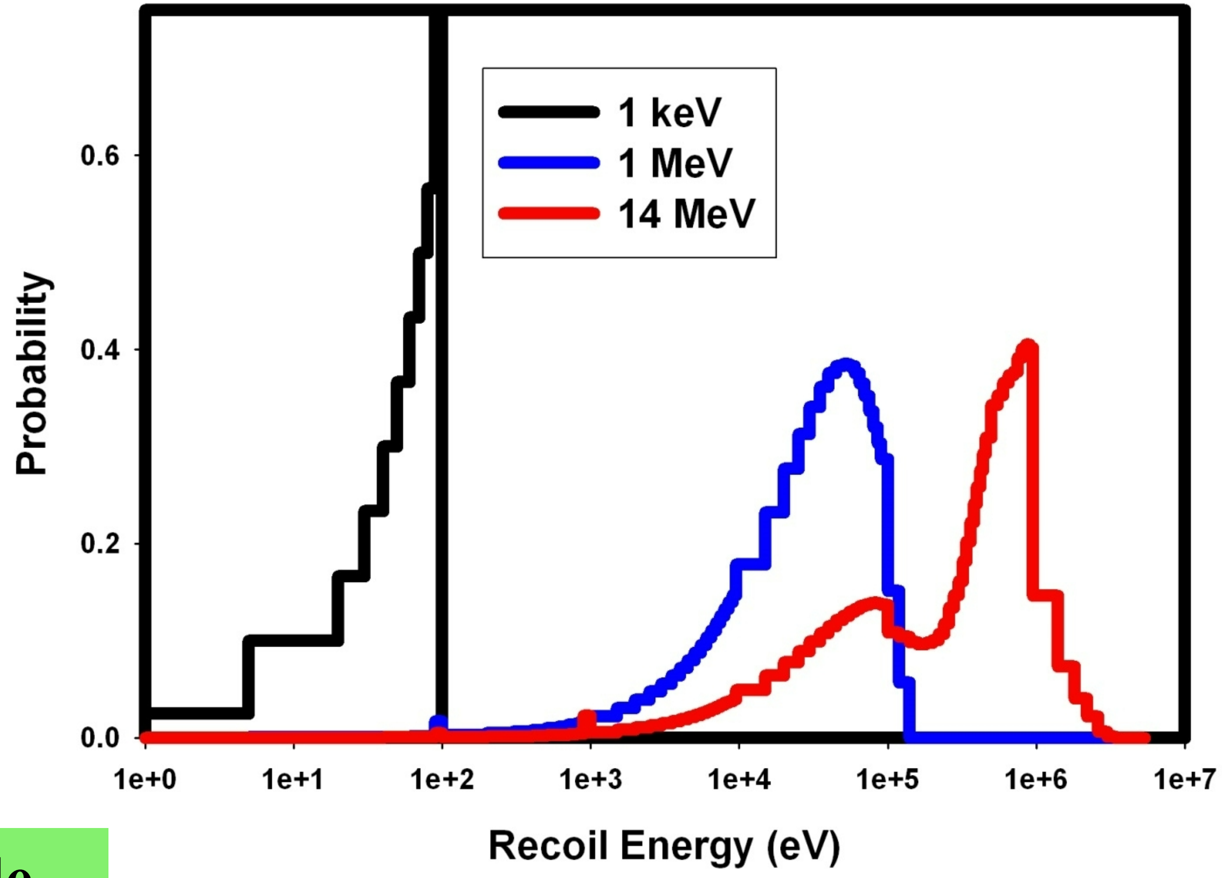
Agenda:

- Purpose
- Terminology
  - Environment vs. Effect
  - Stopping Power vs. LET
- Application space
- Nuclear Data Needs
  - Details of need are coupled with the motivation from the application

Why are "stopping powers" important?

This needs to be understood in the context of the mission!

Neutron-induced Recoil Ion Spectra in Silicon

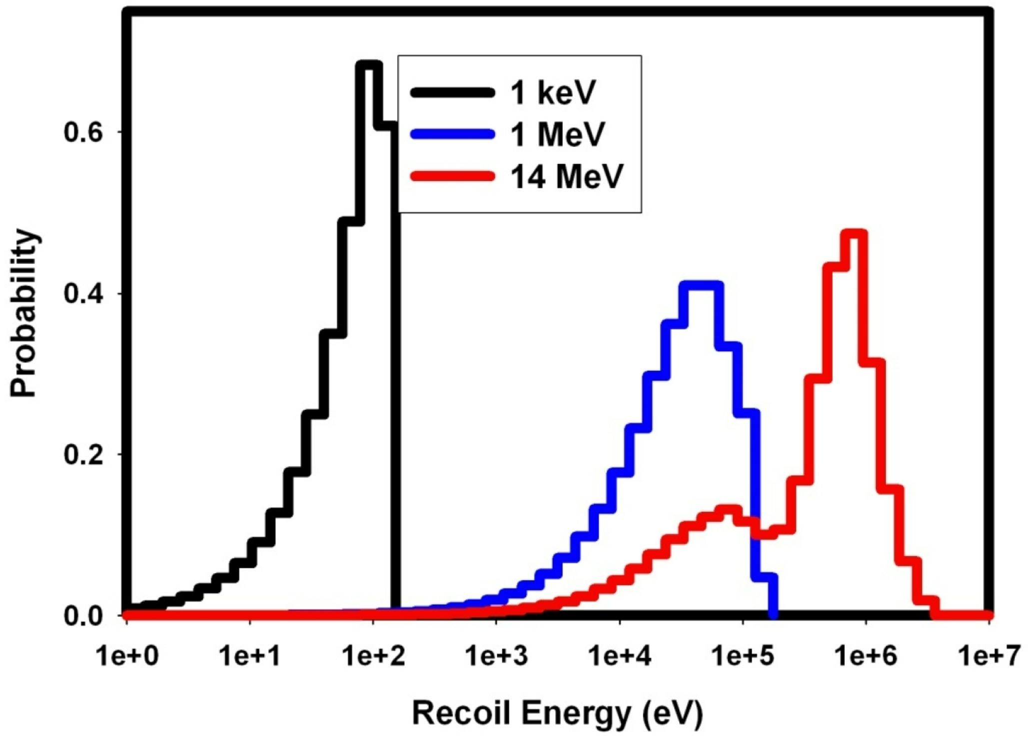


SPECTER module

Strong variation in the recoil ion energy with incident neutron energy.

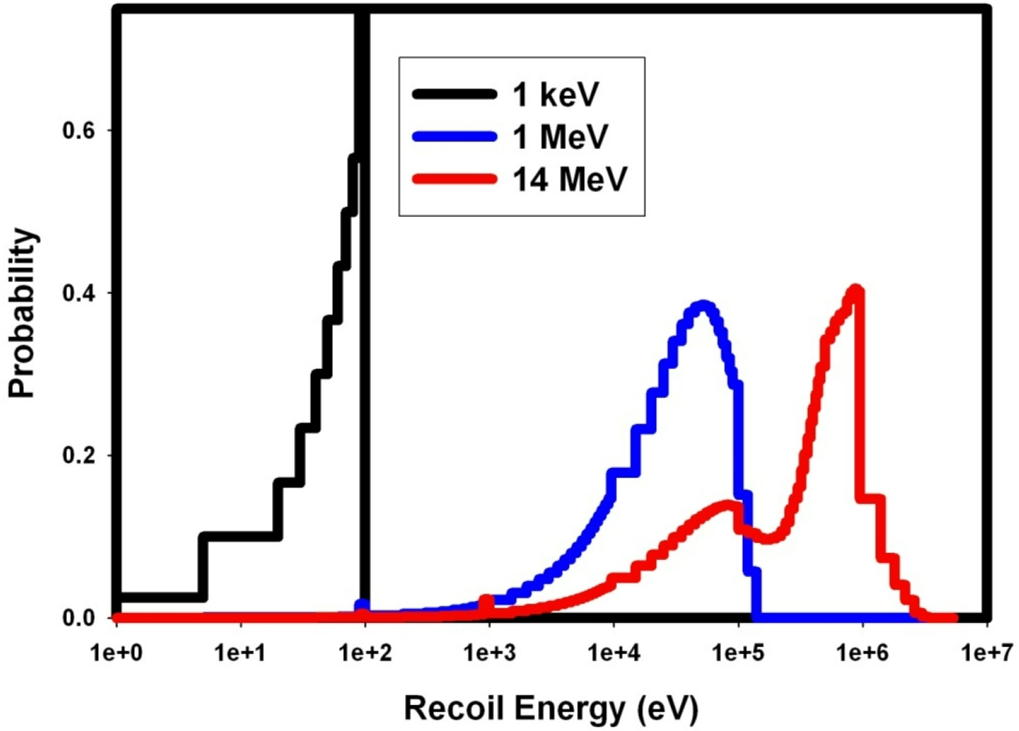
Alternate Models for Si Recoil Spectra

EMPIRE module



$<E>_{1\text{-kev}}$ = 70 eV
$<E>_{1\text{-Mev}}$ = 41 keV
$<E>_{14\text{-Mev}}$ = 569 keV + 5.58 MeV α

SPECTER module

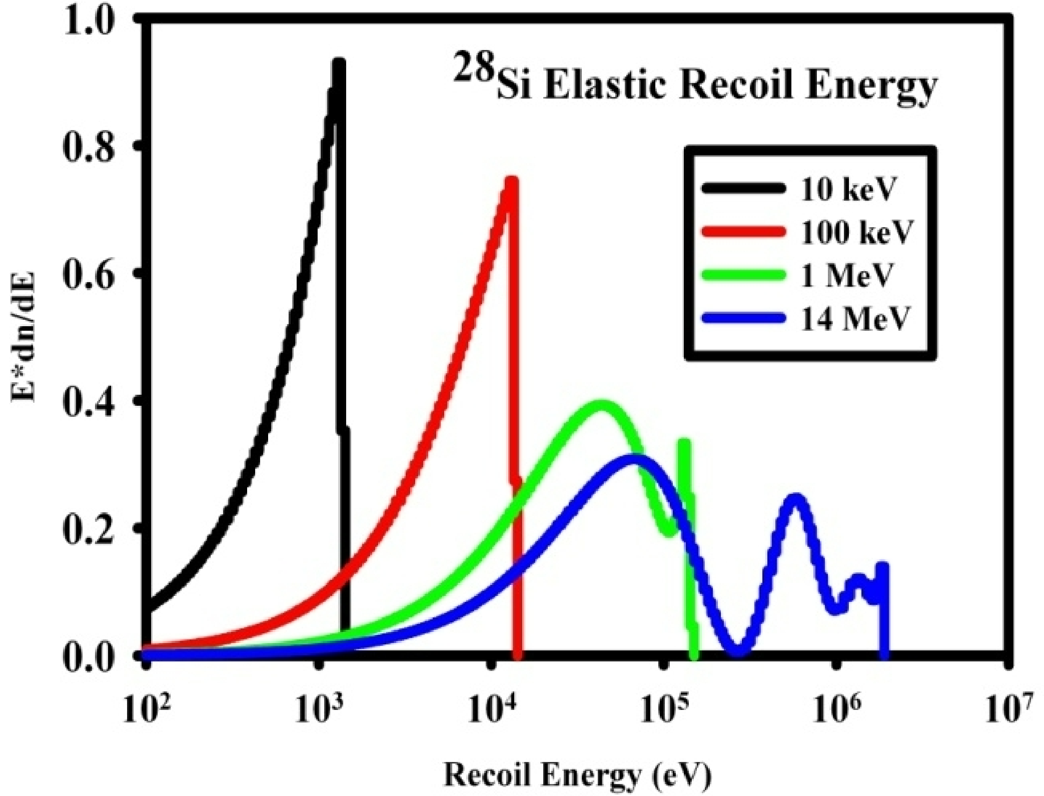


$<E>_{1\text{-kev}}$ = 59 eV
$<E>_{1\text{-Mev}}$ = 39 keV
$<E>_{14\text{-Mev}}$ = 490 keV

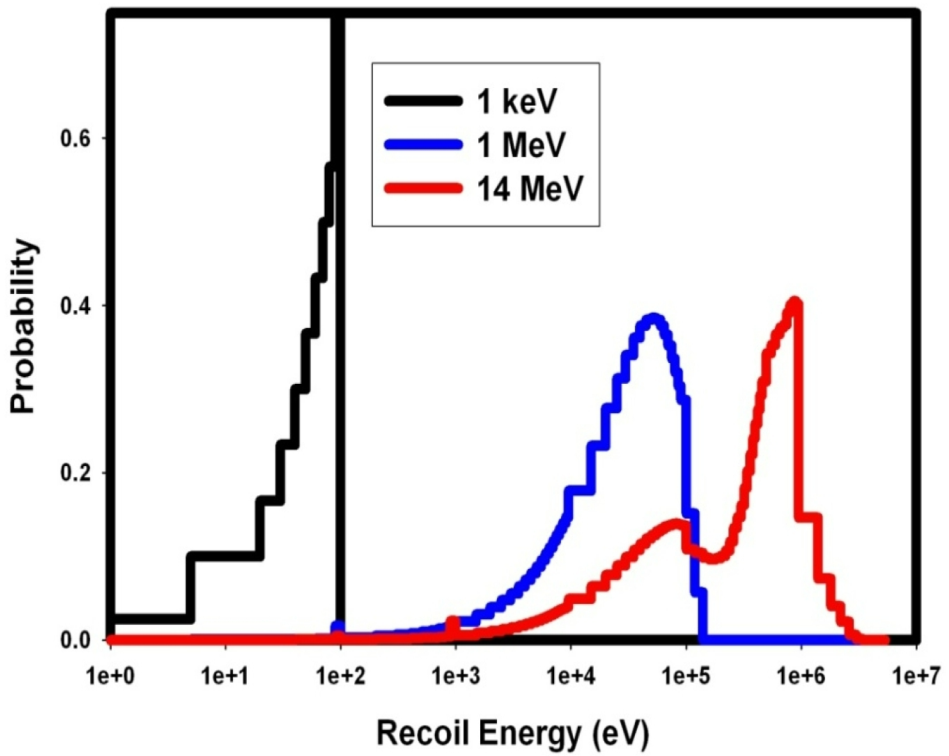
Significant model-based uncertainties exist.

Different Types of Reaction Generate Different Recoil Spectra

Elastic Recoils



PKA Recoils

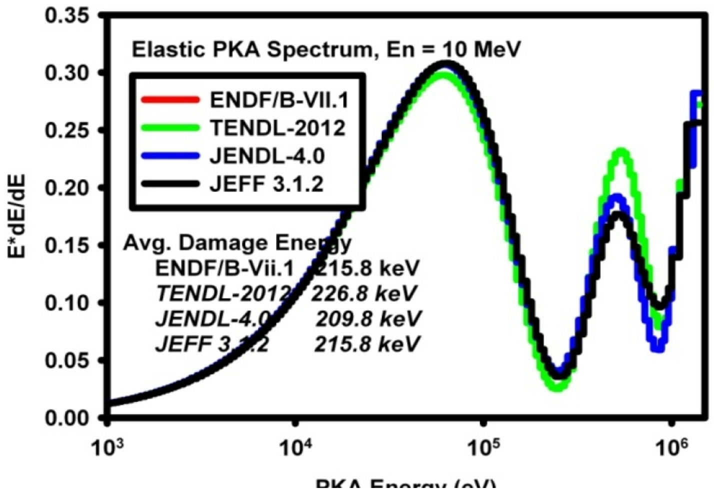


$E_{max\text{-}recoil} = 2A*E_n/(A+1)^2$

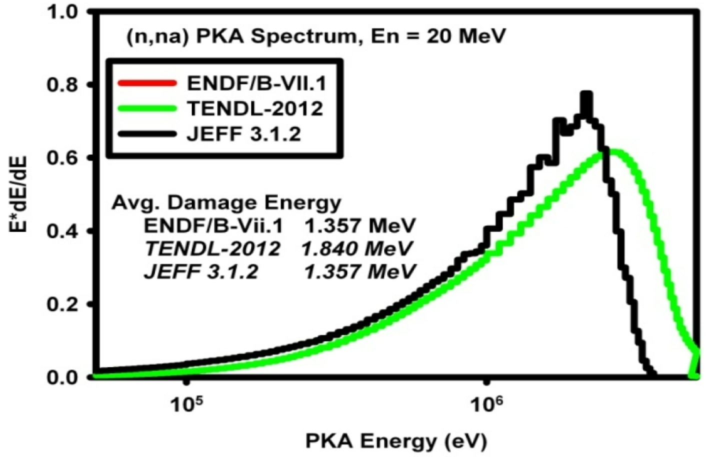
Sharp maximum recoil energy in elastic reactions. Complex recoil spectra for other reactions.

Uncertainty in Silicon Recoil Spectra: Reaction and Energy Dependent

Some Good

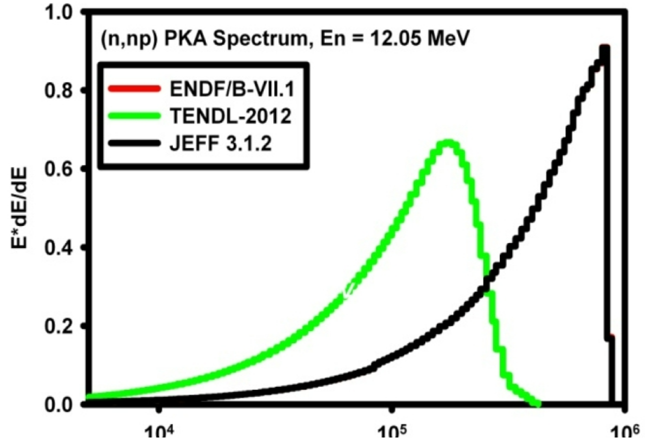


Elastic
10 MeV

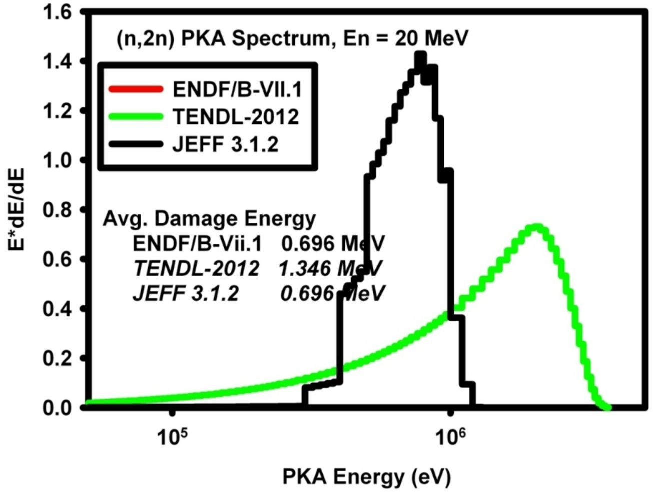


(n,nα)
20 MeV
(E_t = 10.3 MeV)

Some Poor



(n,np)
12.4 MeV
(E_t = 12 MeV)

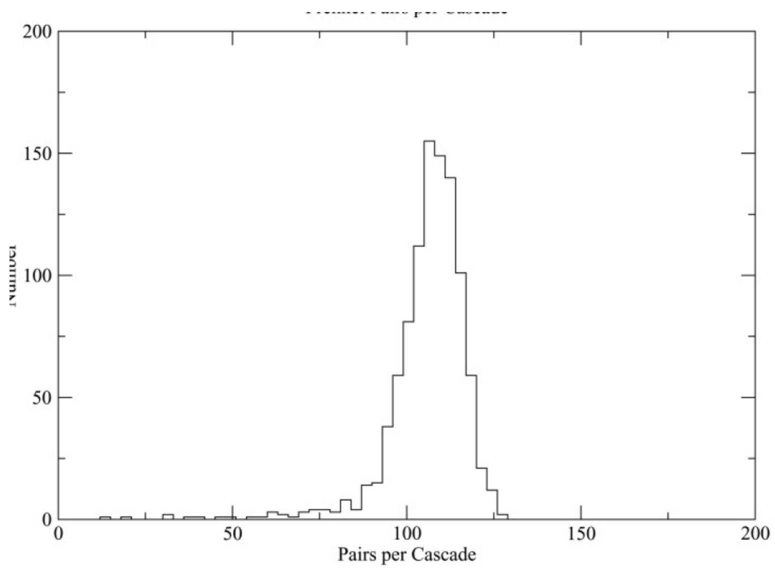


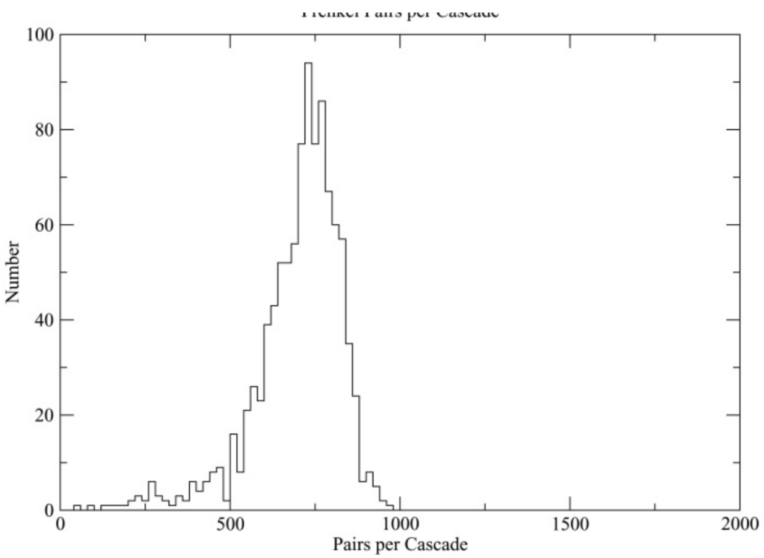
(n,2n)
20 MeV
(E_t = 17.8 MeV)

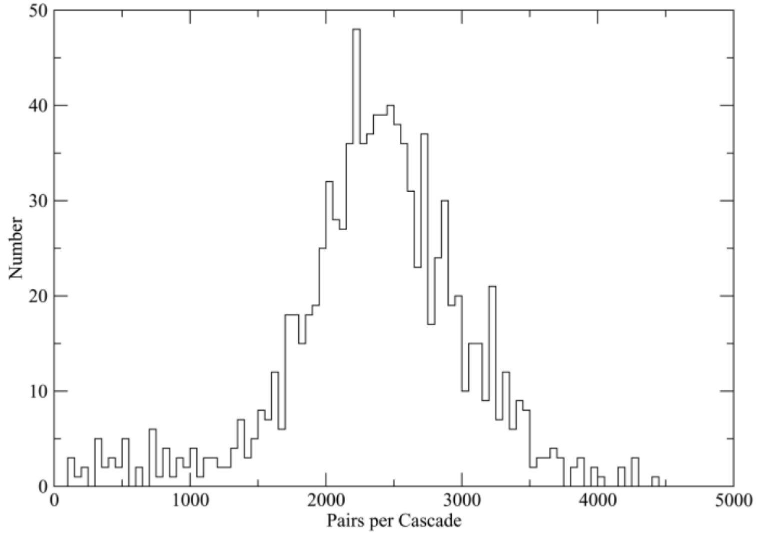
T.

Probability distribution for FP production for a given ion / energy

10 keV	100 keV	1 MeV
<FP> = 103	<FP> = 680	<FP> = 2320
FWHM = 11.5	FWHM = 129	FWHM = 656





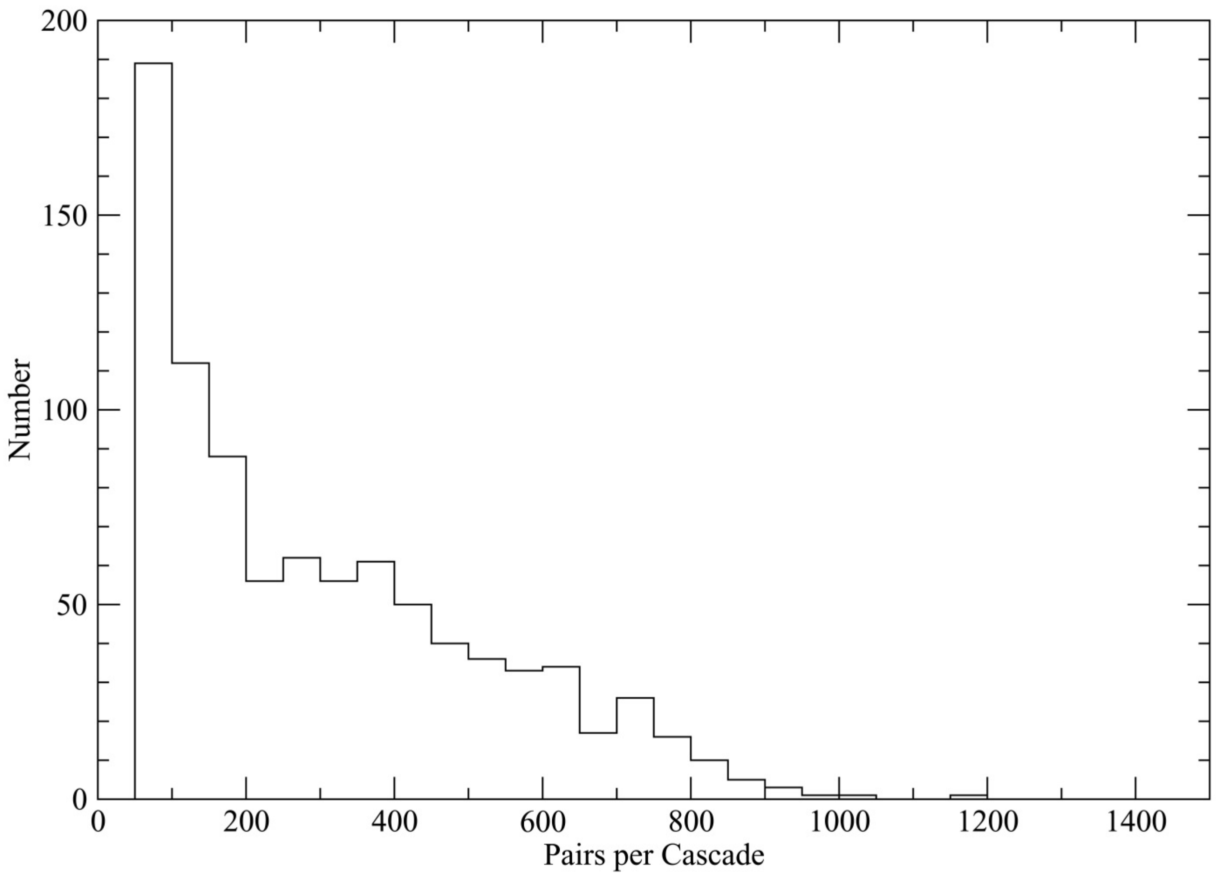


From P. J. Cooper

The variation in the damage track results in significant variation in Frenkel pairs generated.

Now convolve the neutron recoil spectrum with the variation in ion FP generation

pdf for 1-MeV neutrons in silicon lattice.
Sample size = 897 cascades



<FP> = 244
FWHM = 216

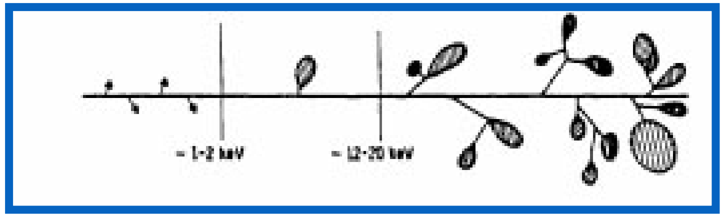
$<E_{recoil}>$ = 38 keV
FWHM = 31 keV

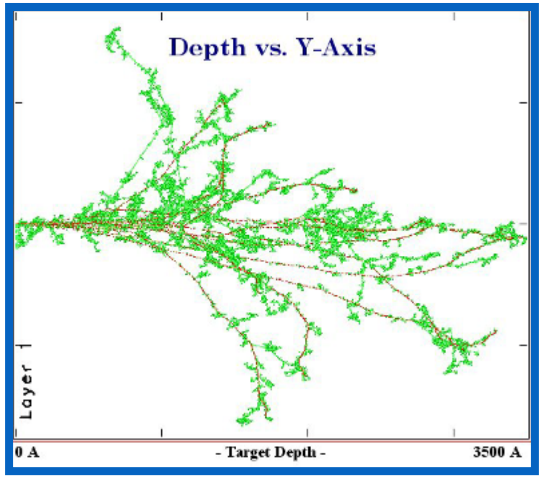
From P. J. Cooper

The distribution is critical!
The variation in the pdf is larger than the mean.

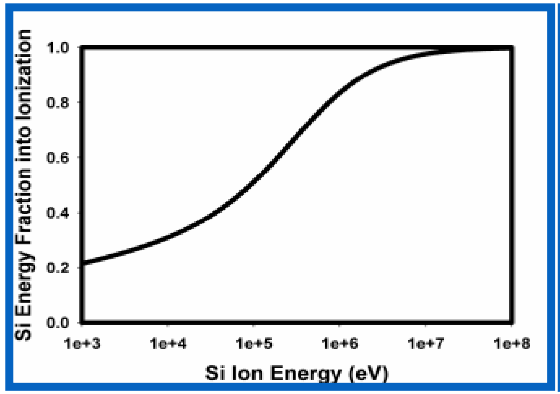
How Ions Interact with Matter

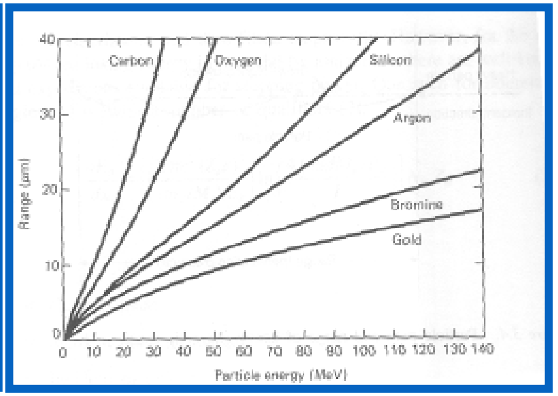
Track Structure

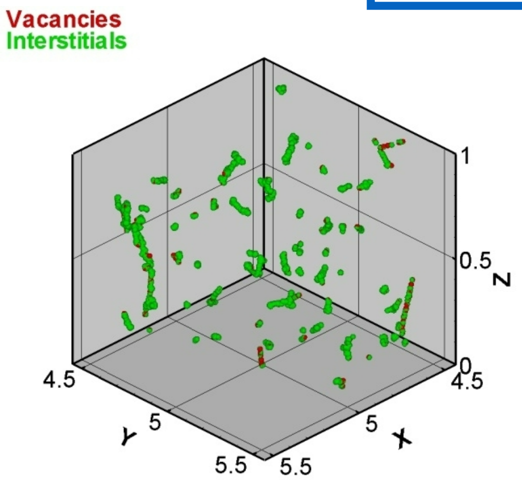


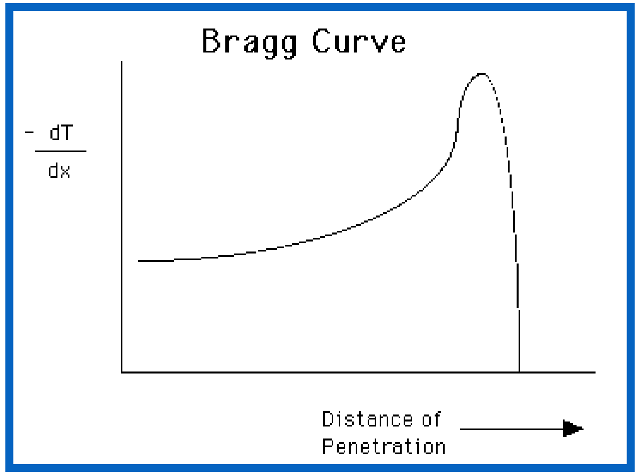


Energy Partition



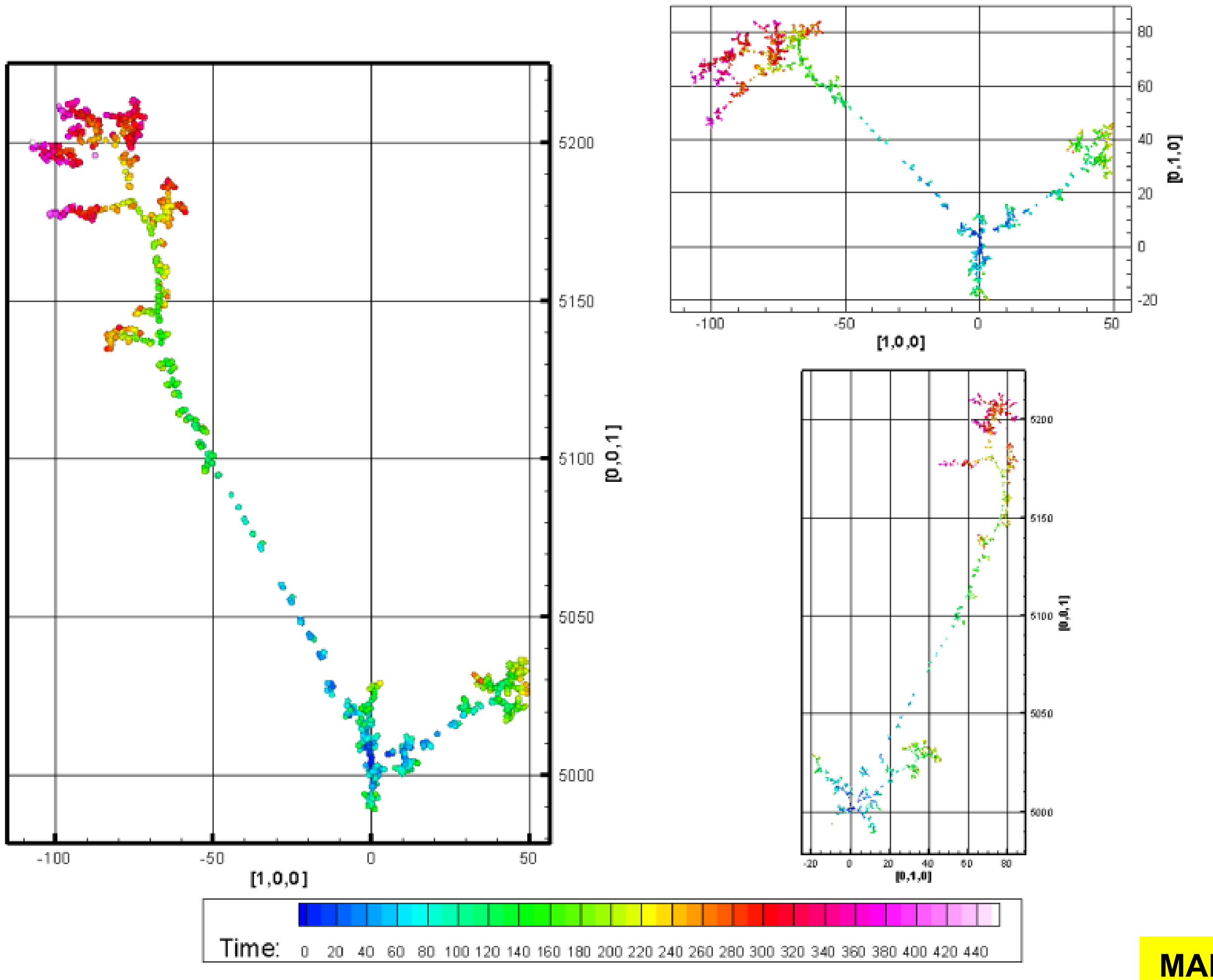






There is a complex structure – in defect generation and in charge release.

Lattice displacements from a single 100-keV Si ion track



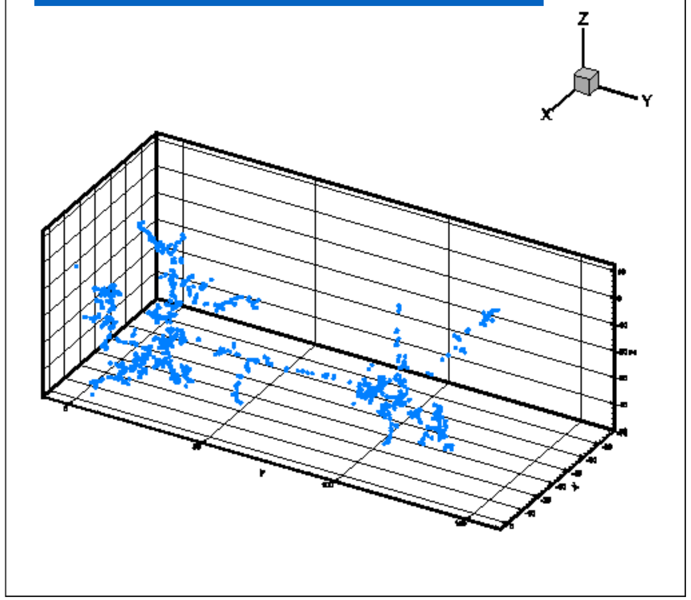
MARLOWE calculation by P. J. Cooper

There is a complex track structure for ion displacement damage. Initial interactions result in both damage clusters and tracks.

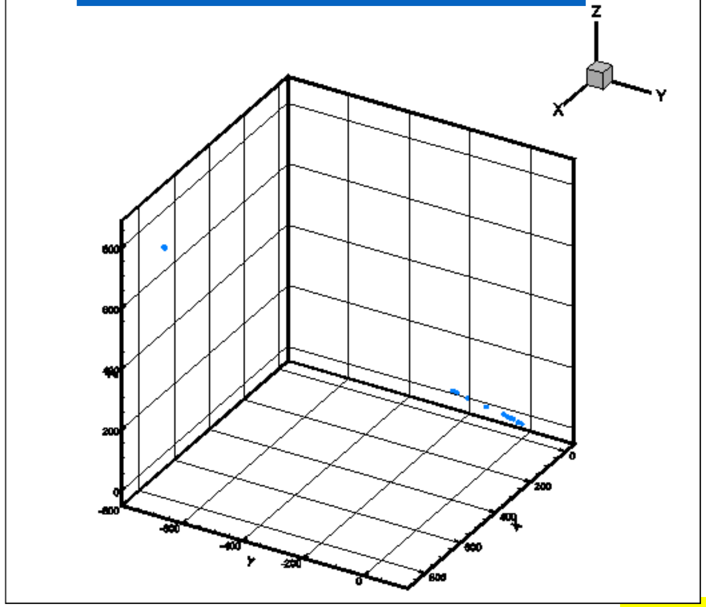
Statistical Process: there is a significant cascade-to-cascade variation in FPs

Two sample cascades for a 100 keV silicon ion in silicon lattice.

925 distant FP
(Vacancies only)



40 distant FP
(Vacancies only)



From P. J. Cooper

For a given incident ion/energy, there are large statistical variations in the damage.

Charge Deposition: Direct Ionization

LET = Linear Energy Transfer

LET is the energy loss per unit path length, normalized by the target material density ⇒ MeV-cm^2/mg

For a given material, LET can be related to the linear charge deposition (LCD) per unit length

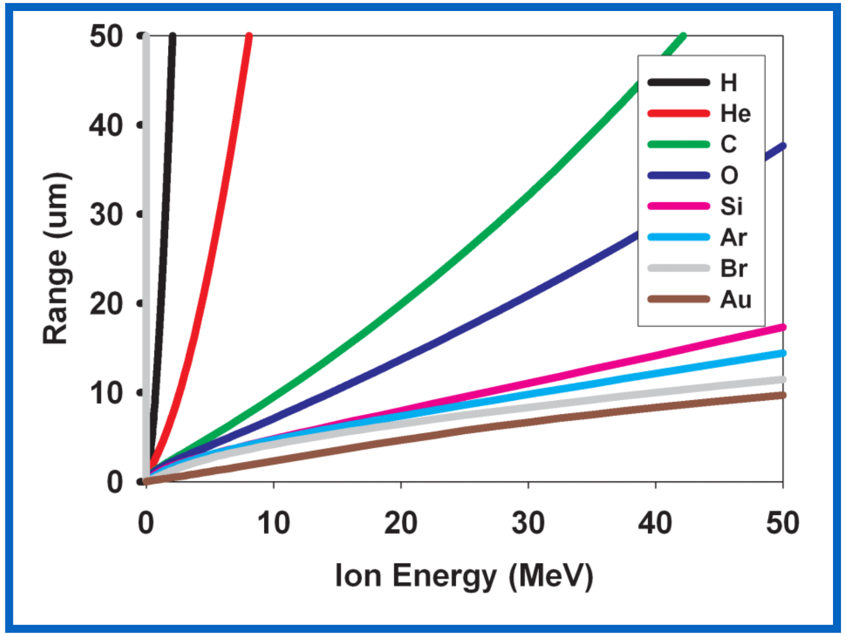
For Si: 97 MeV-cm^2/mg ≈ 1 pC/µm

Direct ionization is the primary charge deposition mechanism for heavy ions (Z ≥ 2).

Source: Paul E. Dodd, HEART short course, 2004.

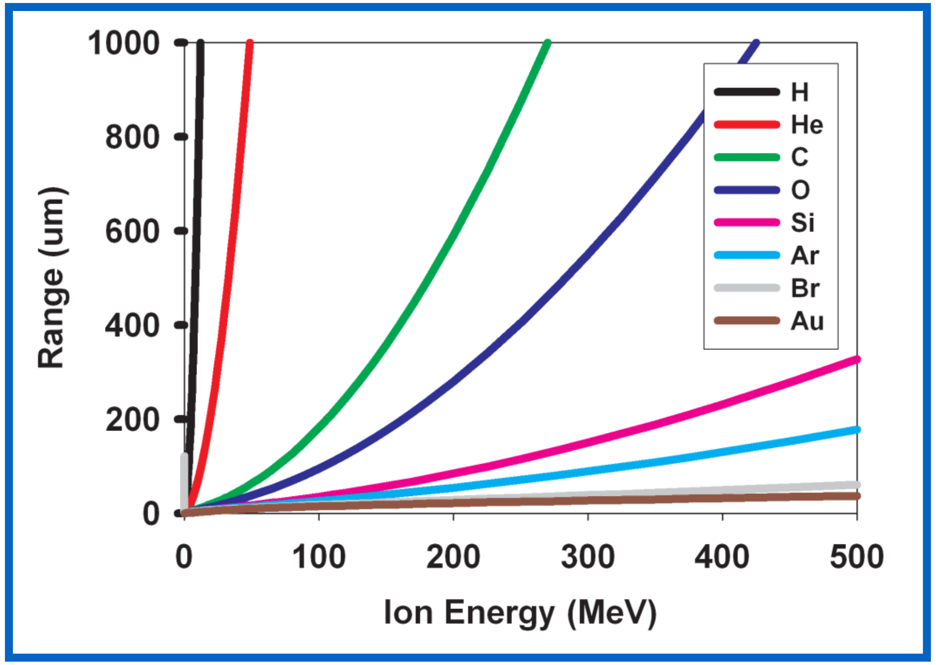
The peak in the stopping power also corresponds to a spatial peak in the energy deposition.

Ion Range in Silicon Lattice



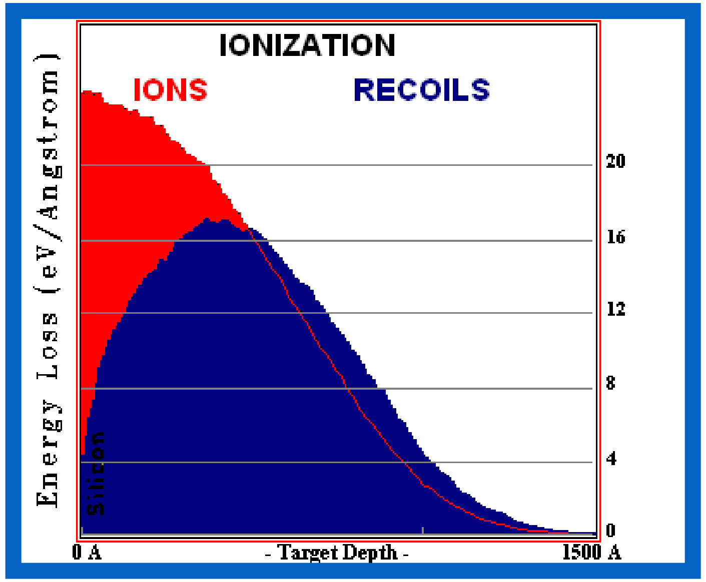
- 5 MeV H has a range of 216 um
- 25 MeV Si has a range of 9.5 um
- 50 MeV Au has a range of 9.7 um

- 12.5 MeV H has a range of 1000 um
- 300 MeV Si has a range of 150 um
- 500 MeV Au has a range of 37 um



Ion range is a limiting consideration in testing packaged electronic parts.

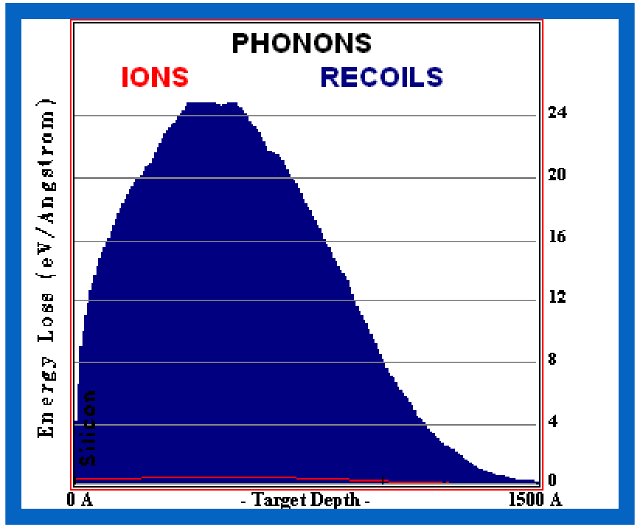
Silicon Recoil Damage



Ionization generation is matched between primary and recoil ion

Energy Loss Mechanism, (50 keV Si ion in Si lattice, range = 736 A, straggle = 290 A)	% Energy Loss	
	Primary Ion	Recoil Atoms
Ionization	30.50	25.67
Vacancies	0.23	3.38
Phonons	0.77	39.44

Phonons generation is dominated by secondary recoils



Deposited is divided into ionization and displacement. Displacement includes bond breaking and lattice phonon generation. Recoils dominate the damage deposition.

n/γ Environment in ACRR Pool-type Reactor Central Cavity

Matl.	Neutron Kerma	Gamma Kerma	% Dose from neutrons
Alanine	469.2	208.2	69%
Diamond	74.3	192.9	28%
Silicon	16.0	203.2	7%
CaF_2:Mn TLD	31.45	202.4	13%

n/γ dose components?

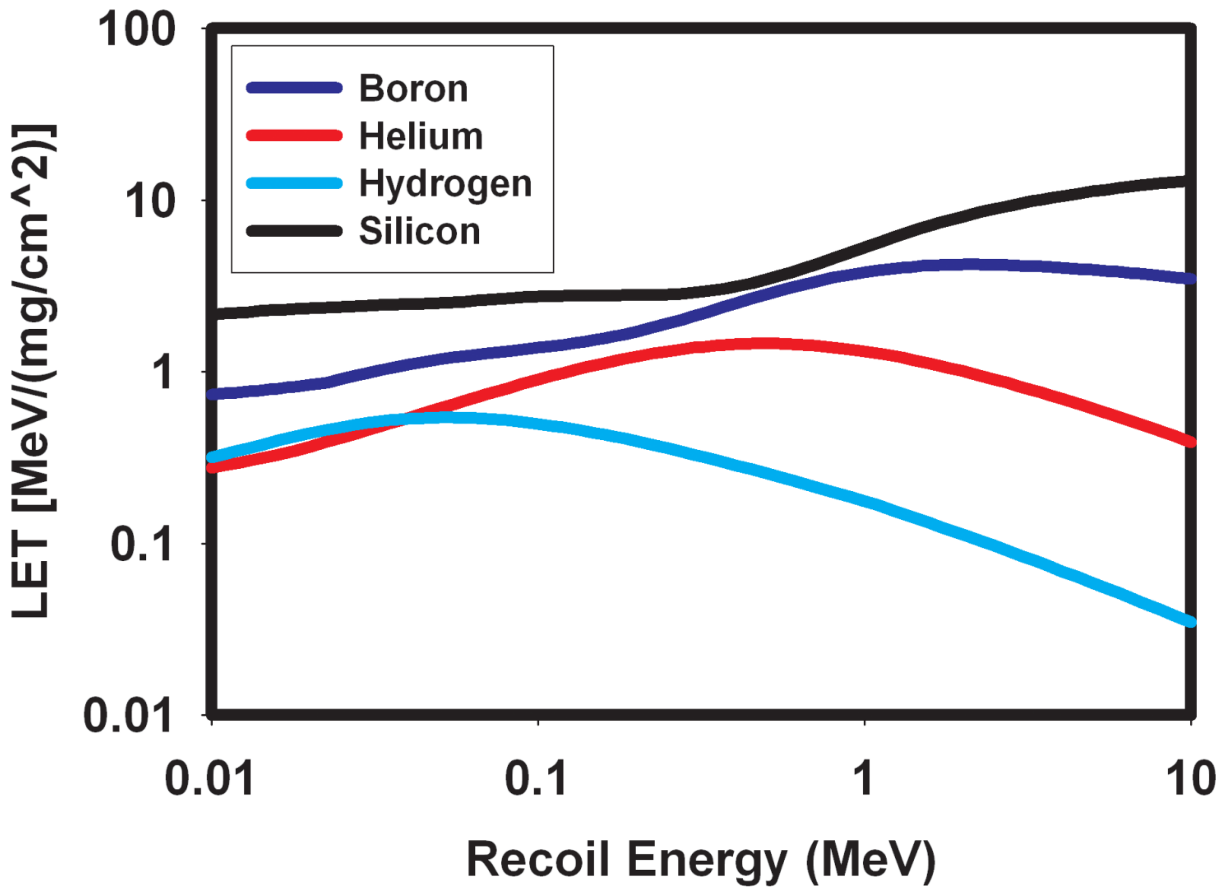
- **photon component varies with Z**
- **neutron component varies with hydrogen content**

Energy Loss Mechanism, (50 keV Si ion in Si lattice, range = 736 A,straggle = 290 A)	% Energy Loss	
	Primary Ion	Recoil Atoms
Ionization	30.50	25.67
Vacancies	0.23	3.38
Phonons	0.77	39.44

Only some of the energy is deposited as ionization!

Ionizing dose in materials in reactor testing must consider both the neutrons and gamma.

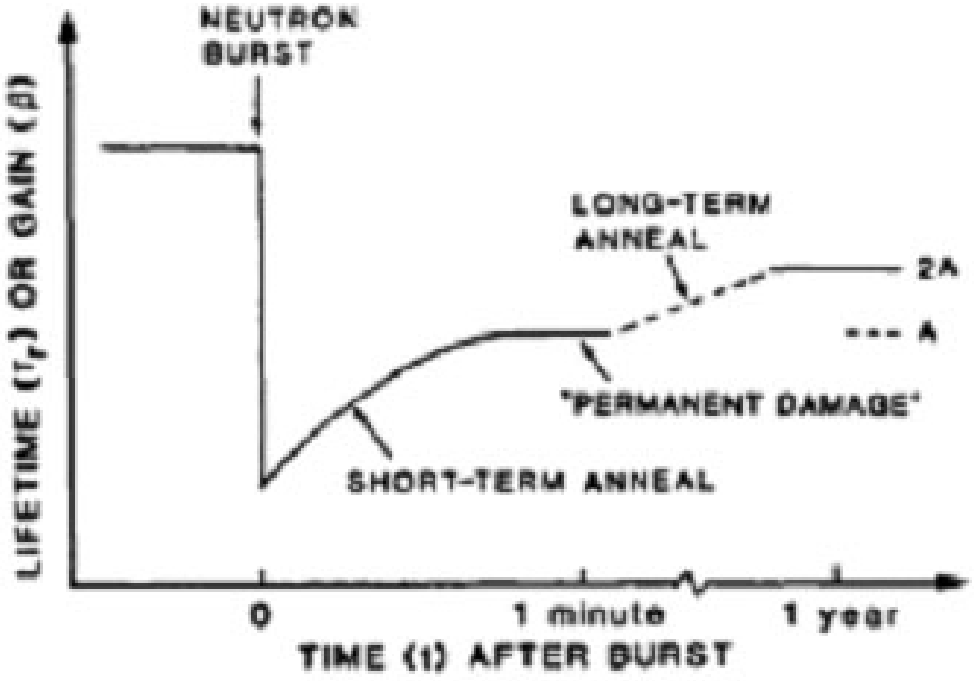
Ion- and Energy-dependent LET in Si (1996 SRIM Stopping Values)

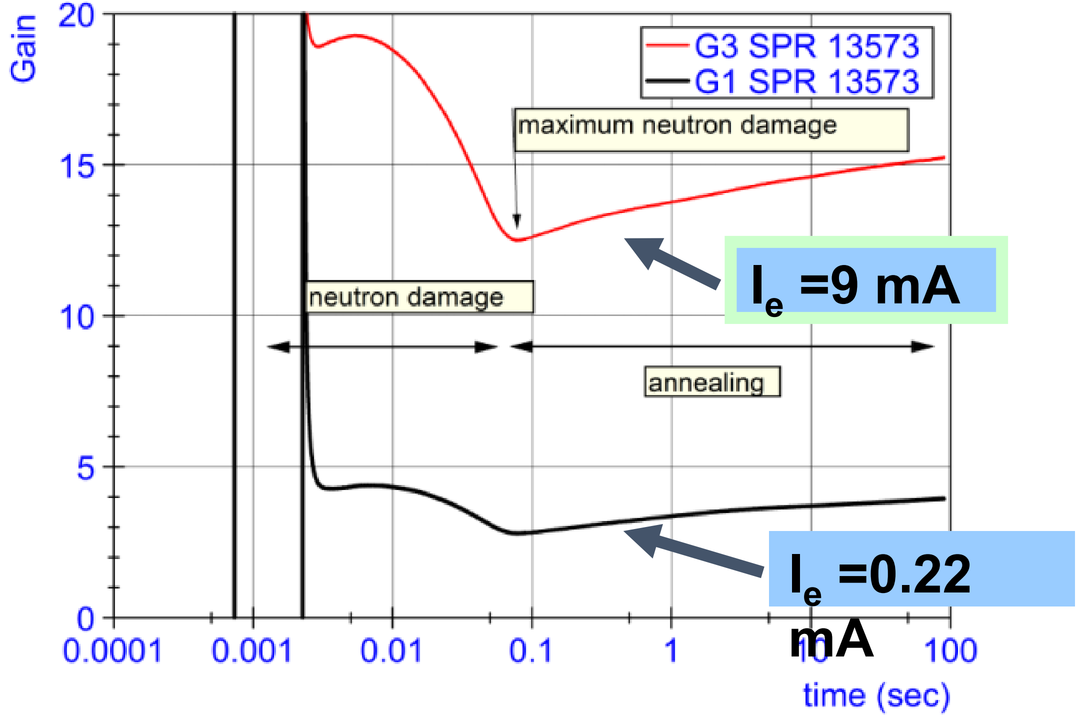


Energy deposited by ionization varies with ion type and energy.

Damage Can Anneal

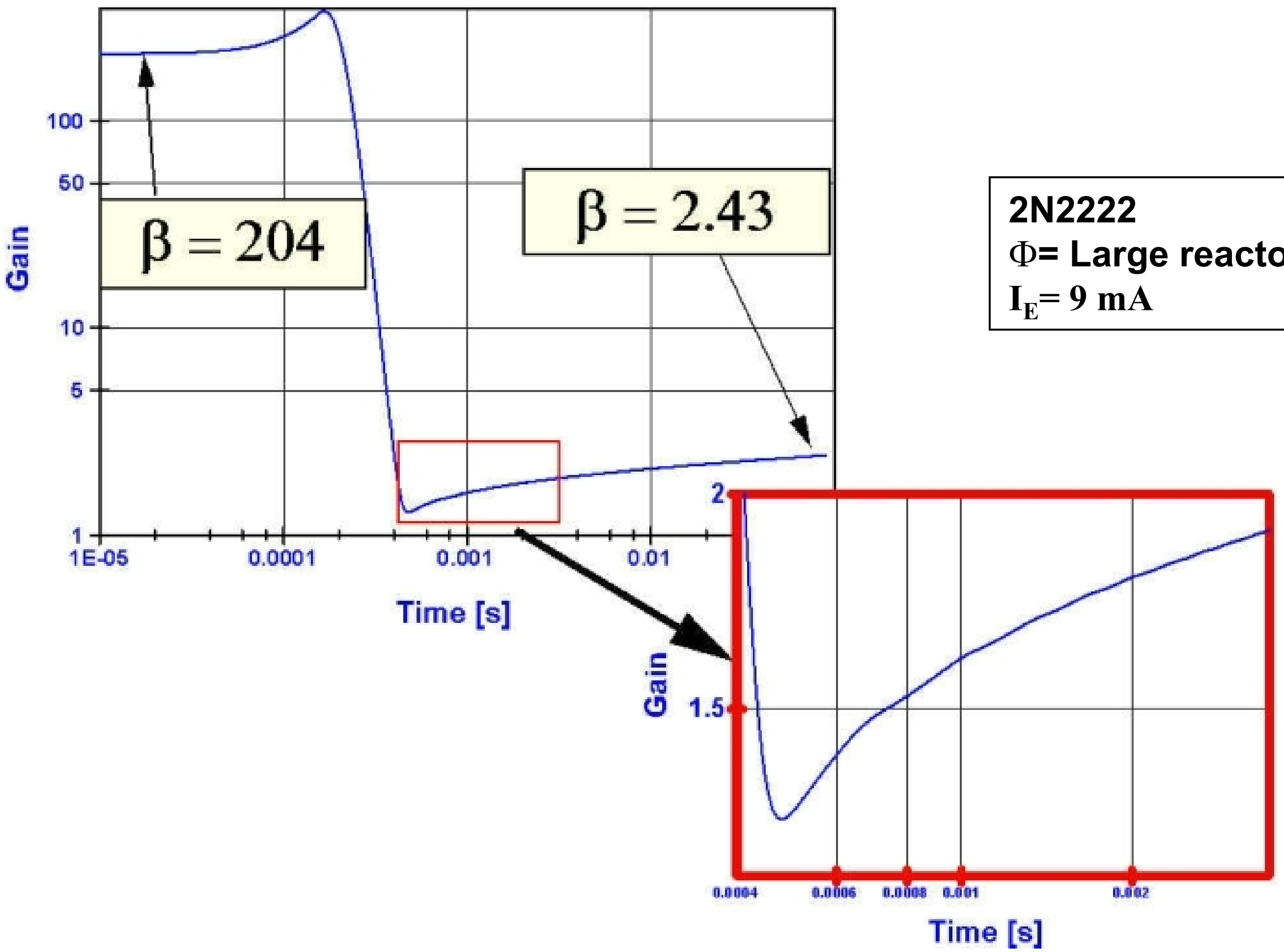
- Different types of annealing:
  - Temperature / time Arrhenius annealing
  - Current injection annealing





Annealing is defect-specific. One must consider the defects that drive damage.

Example of Annealing in 2N2222 Bipolar Transistor



2N2222
Φ= Large reactor pulse
I_E= 9 mA

Photocurrent affects early-time gain. Annealing has several different time constnats due to different defect types that affect the recombination lifetime.

Definition of Annealing Function

Messenger-Spratt Eqn.

$$\left(\frac{1}{H_{fe}(\infty)} - \frac{1}{H_{fe}(0)}\right) = k_n \varphi_n$$

Annealing Function

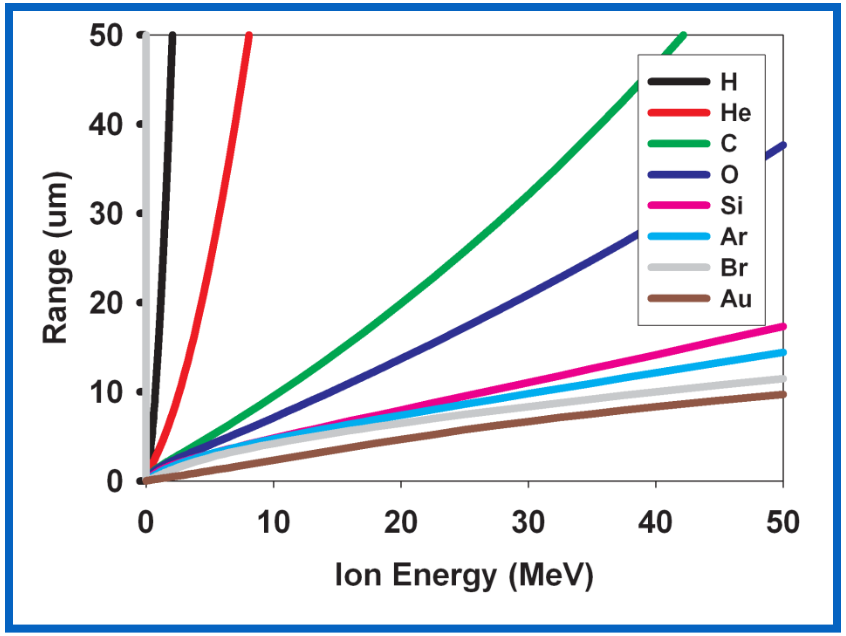
$$\left(\frac{1}{H_{fe}(t)} - \frac{1}{H_{fe}(0)}\right) = AF(t) k_n \varphi_n$$

Normalized Metric

$$AF(t) = \frac{\dfrac{1}{H_{fe}(t)} - \dfrac{1}{H_{fe}(0)}}{\dfrac{1}{H_{fe}(\infty)} - \dfrac{1}{H_{fe}(0)}}$$

Displacement damage is not a linear damage effect.

Ion Range in Silicon



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- 25 MeV Si has a range of 9.5 um
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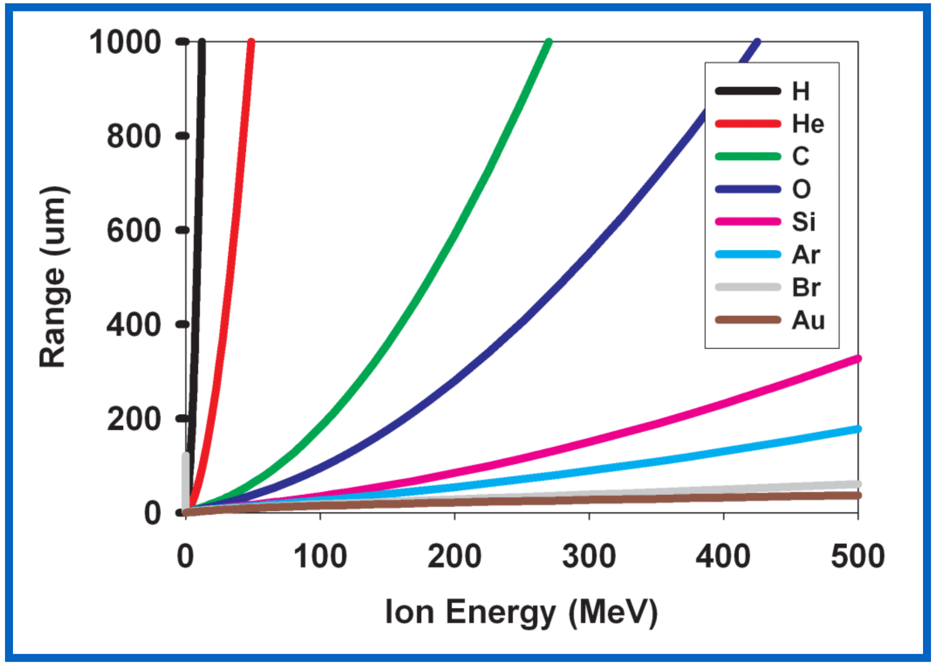


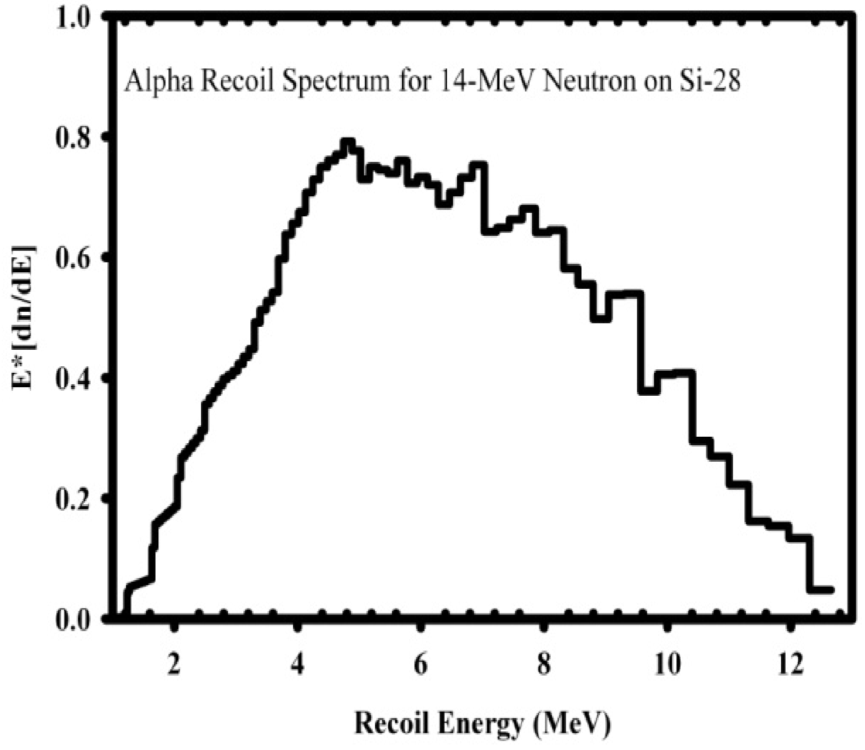
- 12.5 MeV H has a range of 1000 um
- 300 MeV Si has a range of 150 um
- 500 MeV Au has a range of 37 um

Ion energy affects the damage delivered to sensitive volumes in electronics.

Displacement Damage Must Consider All Ions in Outgoing Reaction Channel

Alphas for 14-MeV Neutrons on Si

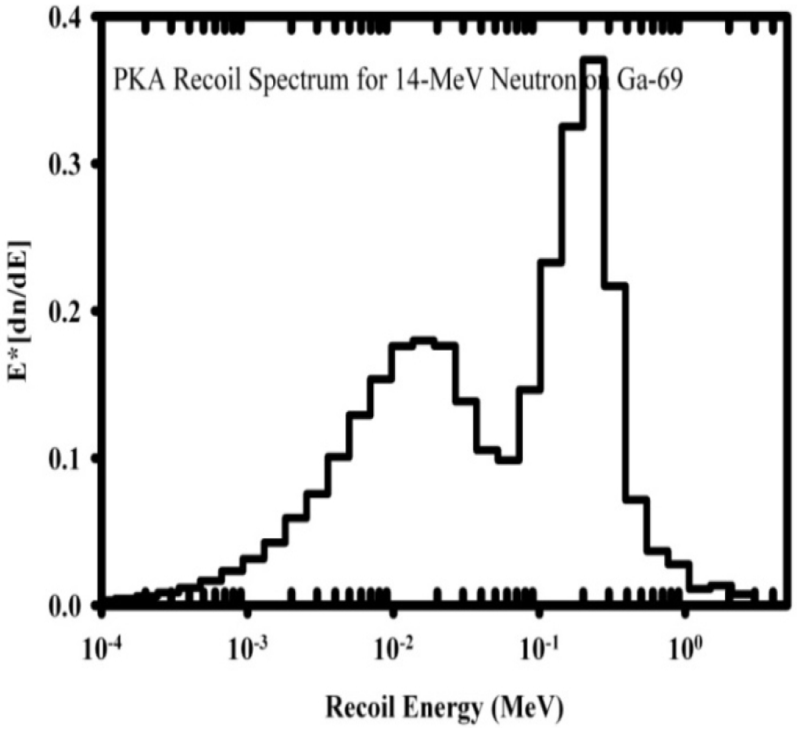
E*dn/dE plot



(n,α); (n,nα); (n,2nα); (n,n2α)

PKA for 14-MeV Neutron on ^{69}Ga

E*dn/dE plot

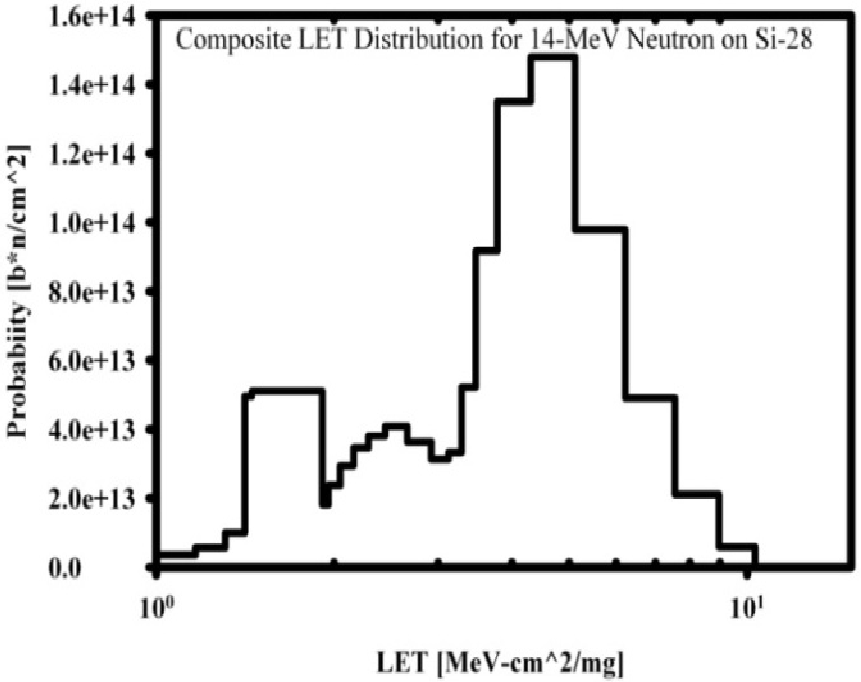


For 14-MeV sources, alpha particle damage must be considered.

Cumulative LET Distributions for 14-MeV Neutrons

^{28}Si

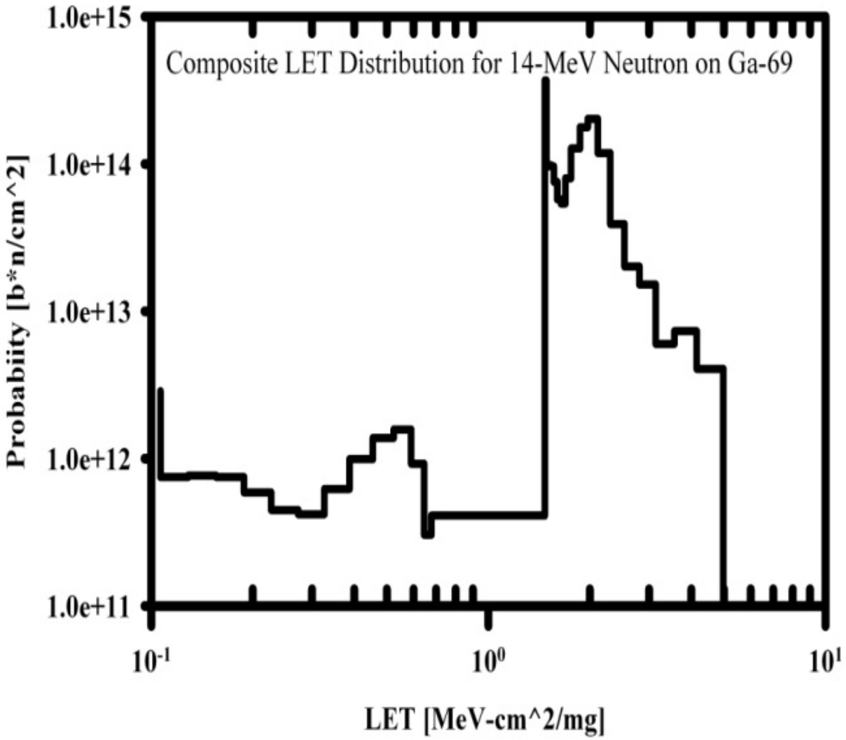
(linear probability axis)



Max. recoil energy = ~10 MeV
Max. LET = ~15 MeV-cm^2/mg

^{69}Ga

(log probability axis)



Max. recoil energy = ~4 MeV
Max. LET = ~5 MeV-cm^2/mg

The LET spectra varies with the material.

End of Backup Material