SAND2017-7102C

Vacancy and Mass-Impurity Phonon Scattering in Self-Irradiated Silicon

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Introduction

- Goal: Irradiate intrinsic Si with different isotopes to experimentally demonstrate the individual effects of mass-impurity and vacancy phonon scattering
  - Phonon: “Quantum of elastic waves” –Tamm, 1930 / Frenkel, 1932

- Si: second most abundant element in Earth’s crust, ubiquitous in electronics, thermal properties thoroughly characterized

- Stable Si isotopes:

Isotope	Prevalence (%)
^{28}Si	92.22
^{29}Si	4.69
^{30}Si	3.09

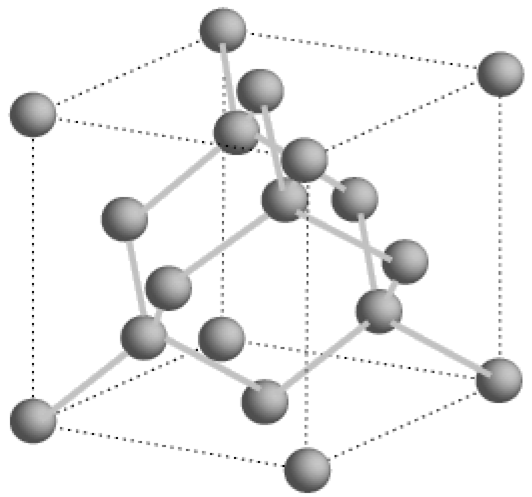
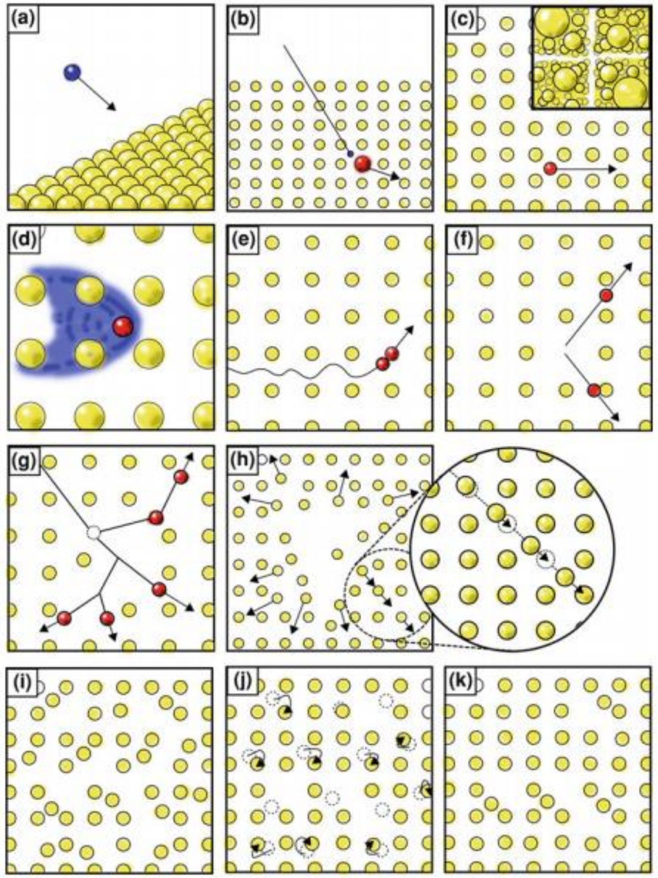
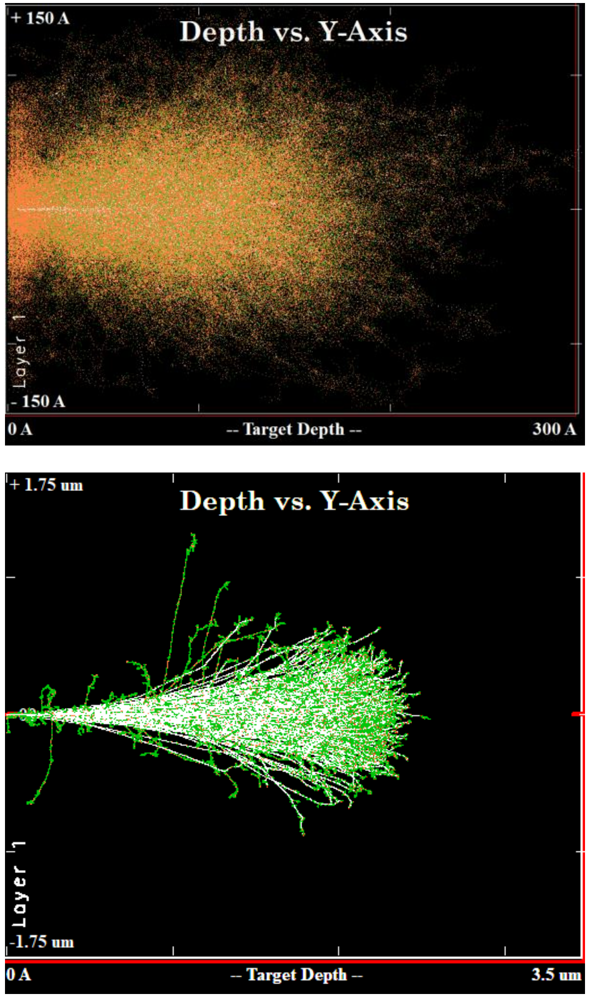


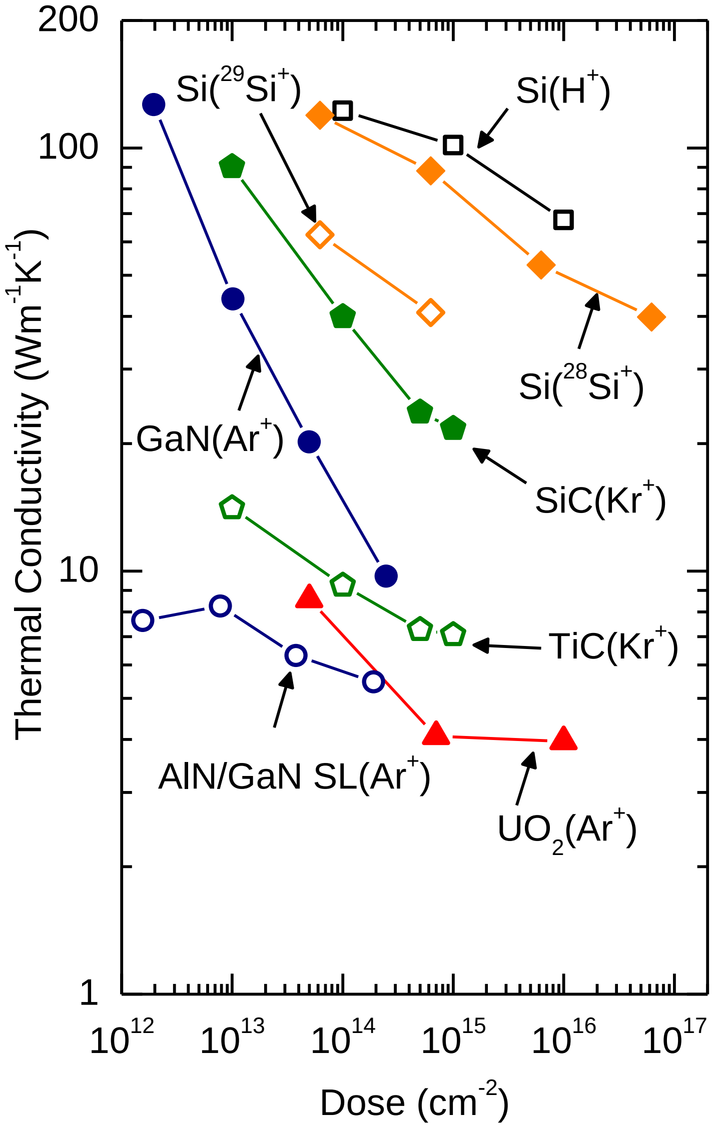
Image source: http://hyperphysics.phy-astr.gsu.edu/hbase/Solids/sili2.html

Ion Irradiation



Race, 2010)Race, C. (2010). The Modelling of Radiation Damage in Metals Using Ehrenfest Dynamics, 309. https://doi.org/10.1007/978-3-642-15439-3





^{28}Si, ^{29}Si Irradiation

- Two sample sets:
  - ^{28}Si – Irradiated $Si_{intrinsic}$
    - Beam Energy: 3.75 [MeV]
    - Dose: 6.24E13, 6.24E14, 6.24E15, 6.24E16 [cm^{-2}]
    - Purpose: Primarily *vacancy* scattering
  - ^{29}Si – Irradiated $Si_{intrinsic}$
    - Beam energy: 3.75 [MeV]
    - Dose: 6.24E13, 6.24E14, 1.47E14 [cm^{-2}]
    - Purpose: add in mass-impurity scattering
- Irradiation: 6 MV Tandem Van de Graaf-Pelletron Accelerator, Sandia National Laboratories

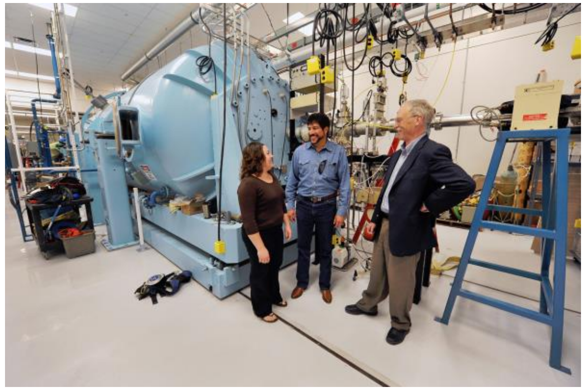
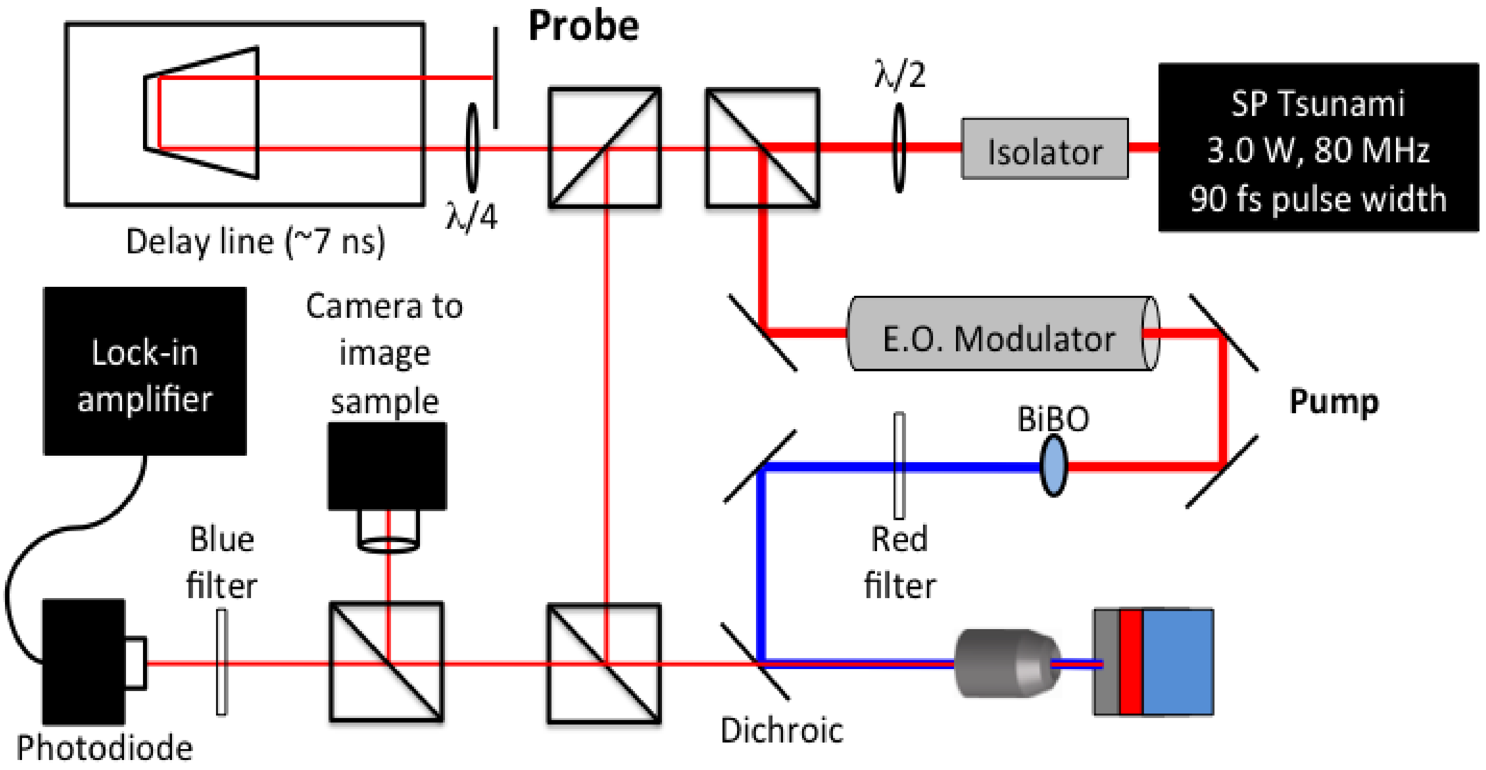


Image source: http://www.sandia.gov/LabNews/100521.html

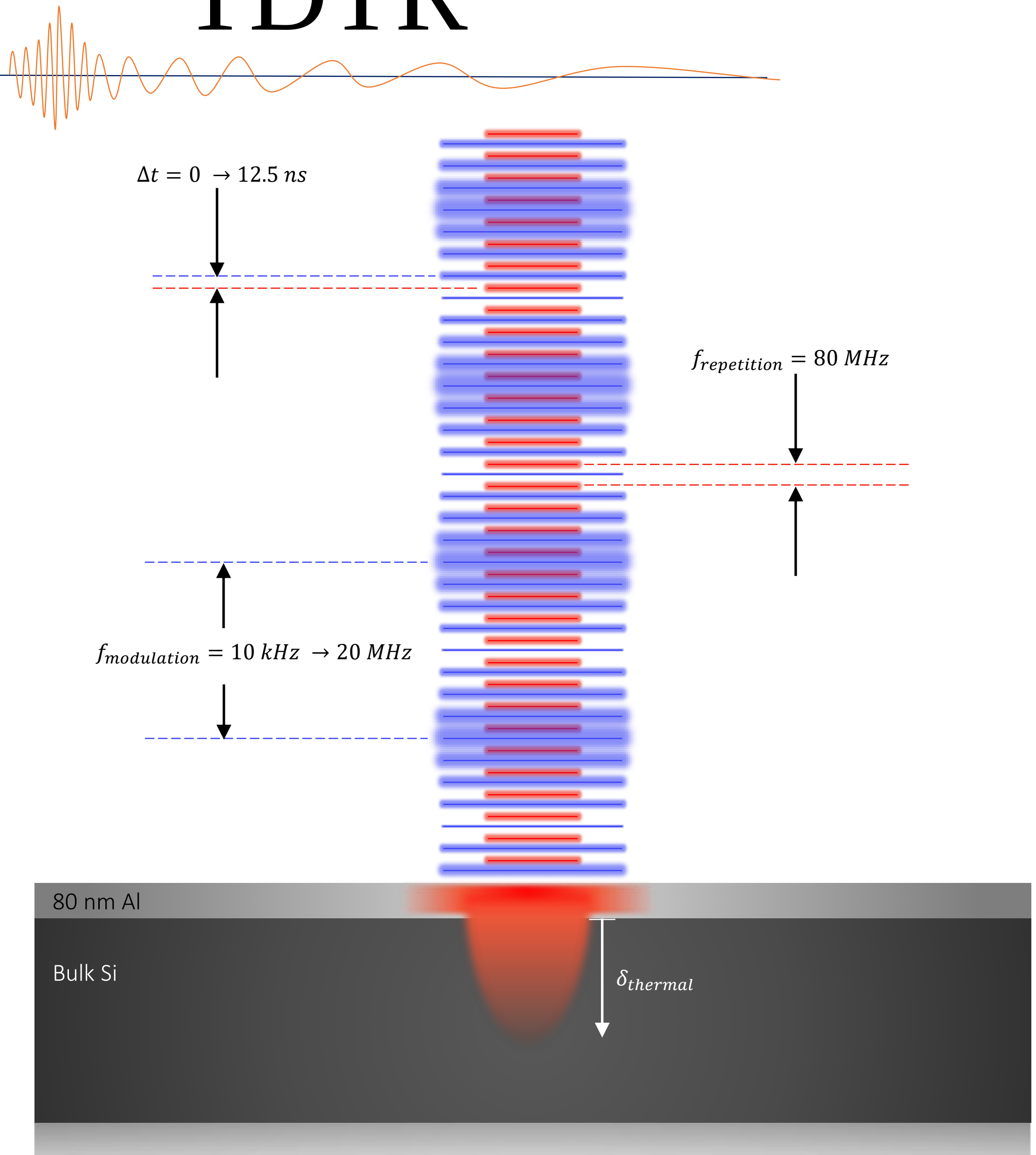
Time-Domain Thermoreflectance (TDTR)

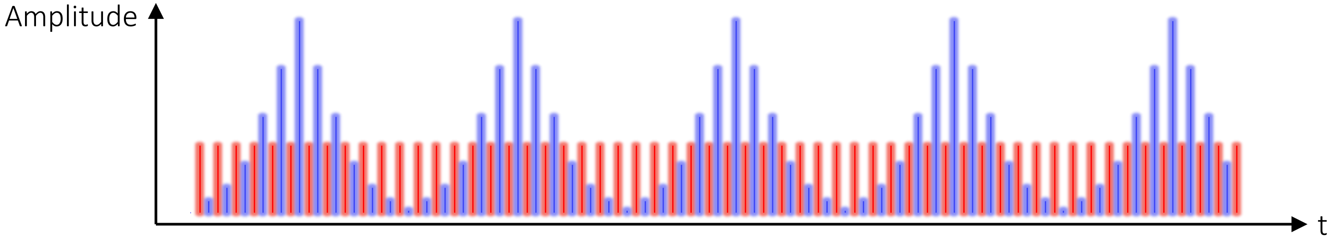


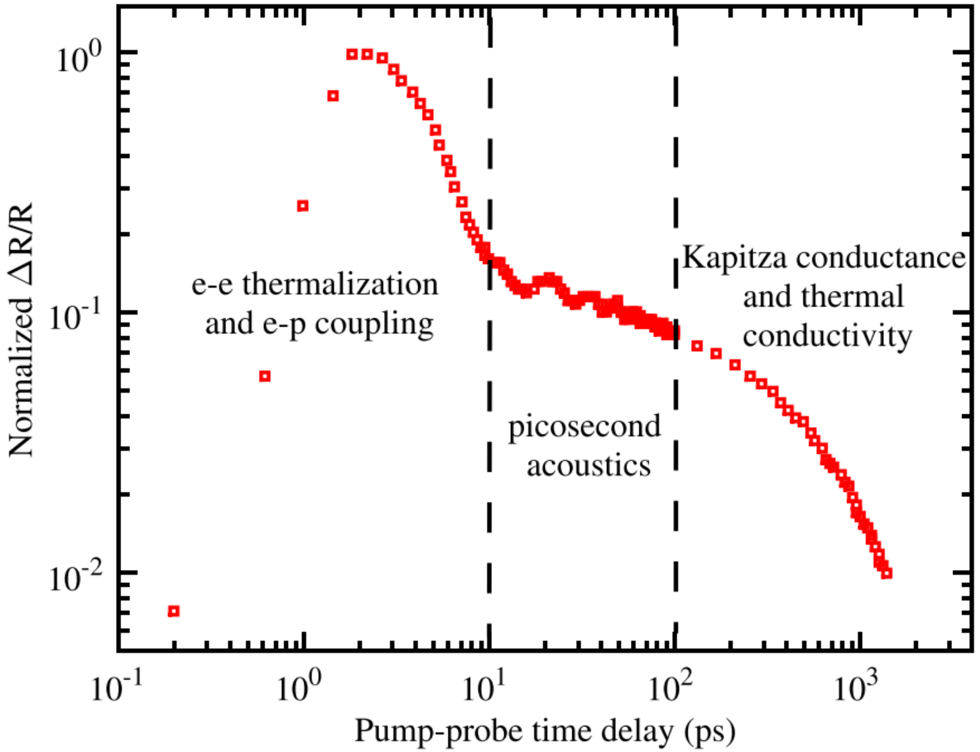
Hopkins *et al.*, *ISRN Mechanical Engineering, 682586* (2013)

- Non-destructive, pump-probe technique
- Sub-picosecond temporal resolution
- Spot sizes as low as ~ 1 µm
- Modulation frequencies: 10 kHz – 20 MHz

TDTR

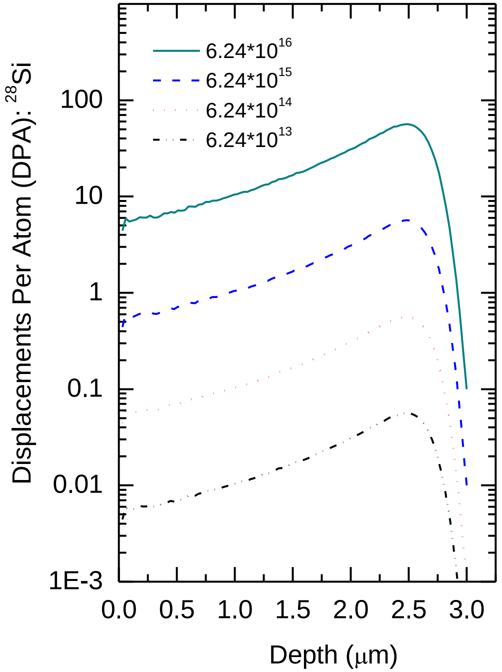




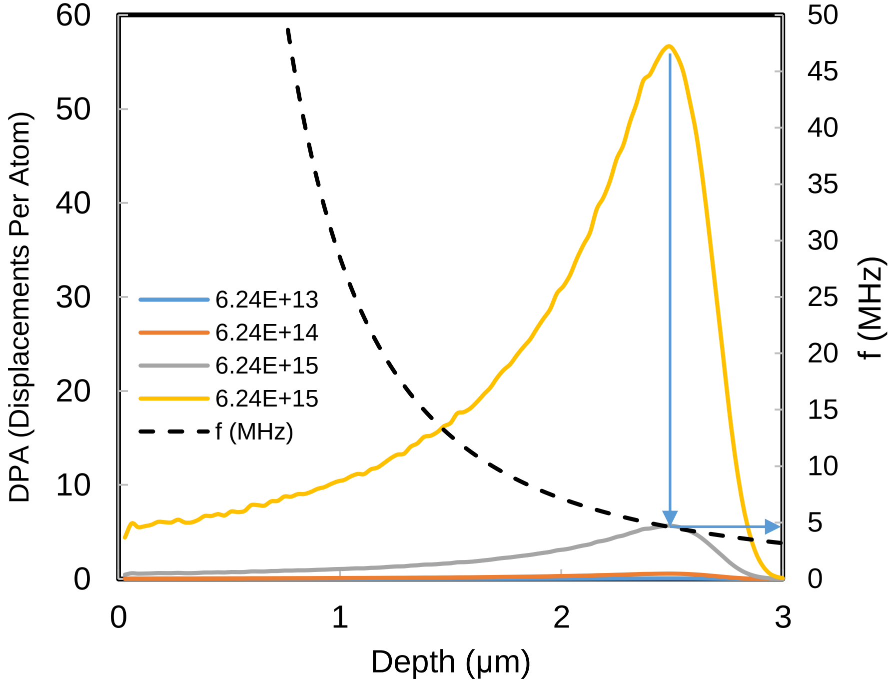


Giri *et al.*, *Journal of Applied Physics*. 117, 105105 (2015)

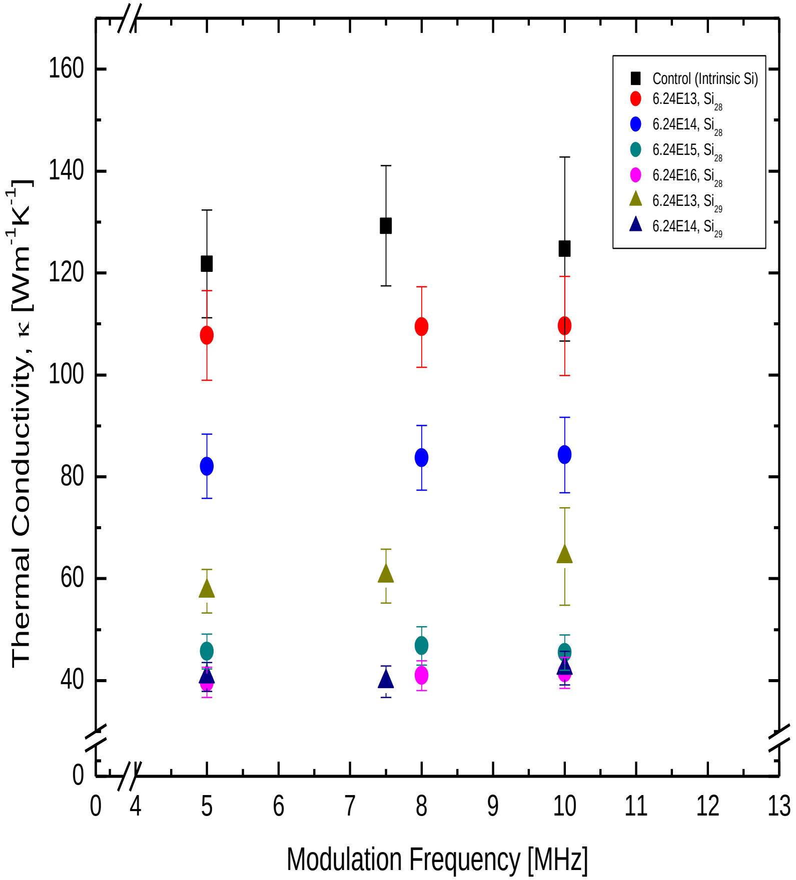
^{28}Si, ^{29}Si Measurement



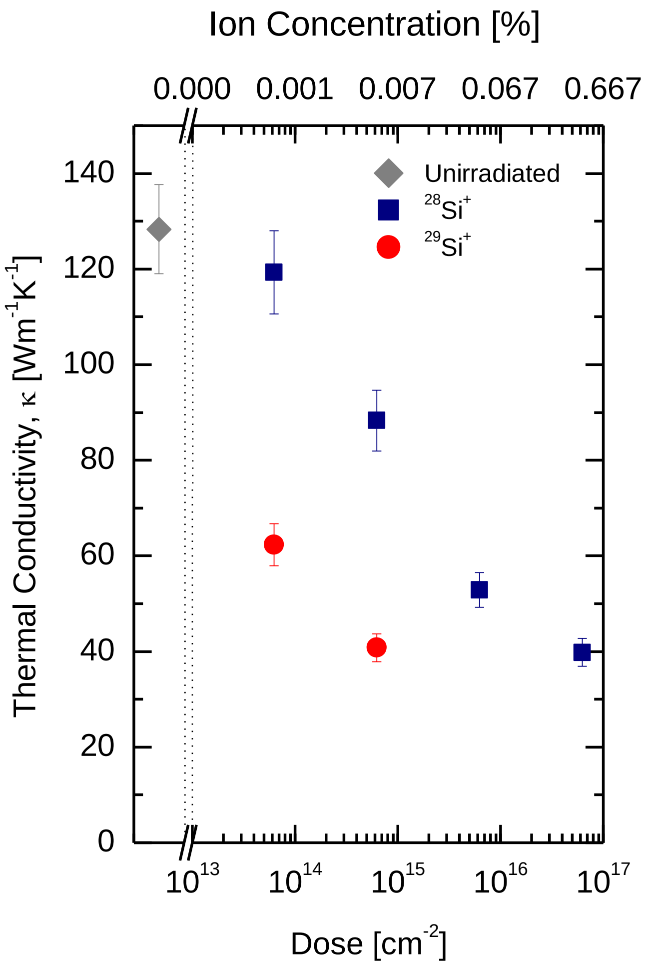
$$\delta_{thermal} = \sqrt{\frac{\kappa}{\pi C f}}$$



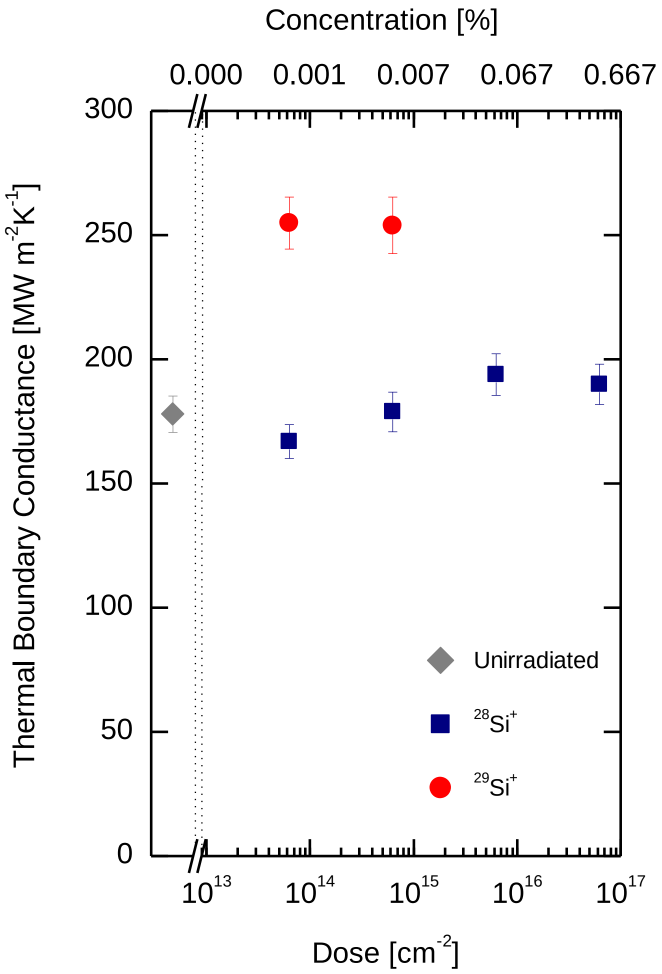
Results



Control (Intrinsic Si)
6.24E13, Si28
6.24E14, Si28
6.24E15, Si28
6.24E16, Si28
6.24E13, Si29
6.24E14, Si29
Thermal Conductivity, κ [Wm-1K-1]
Modulation Frequency [MHz]



Ion Concentration [%]
0.000 0.001 0.007 0.067 0.667
Unirradiated
28Si+
29Si+
Thermal Conductivity, κ [Wm-1K-1]
Dose [cm-2]



Concentration [%]
0.000 0.001 0.007 0.067 0.667
Unirradiated
28Si+
29Si+
Thermal Boundary Conductance [MW m-2K-1]
Dose [cm-2]

Thermal Conductivity

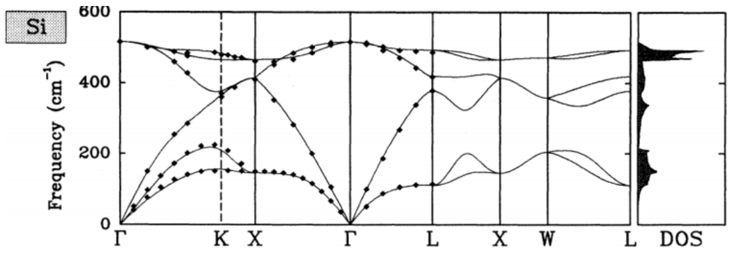
- From kinetic theory:

$$\kappa = \frac{1}{3} C_v v l = \frac{1}{3} C_v v^2 \tau$$

$$\kappa = \frac{1}{6\pi^2} \sum_j \int \hbar\omega_j(k) k^2 \frac{\partial f}{\partial T} v_j^2(k) \tau_j(k) dk$$

- All parameters can be assumed from literature dispersion of Si

- Scattering time, τ is unique from sample to sample



P. Giannozzi, S. De Gironcoli, P. Pavone, and S. Baroni, Phys. Rev. B **43**, 7231 (1991).

First, determine baseline scattering:

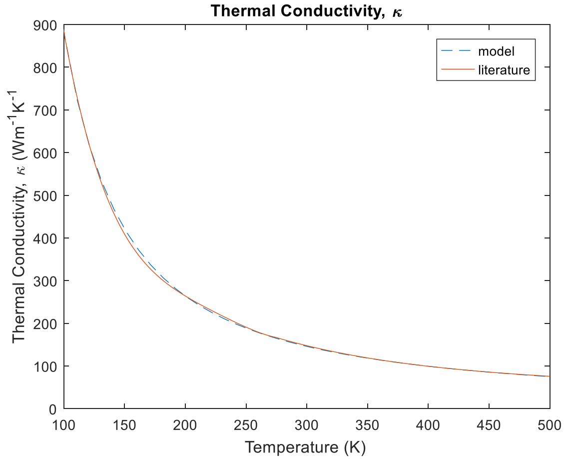
$$\frac{1}{\tau} = \frac{1}{\tau_{Ph}} + \frac{1}{\tau_{Imp}}$$

- Phonon-phonon (Umklapp):

$$\tau_{ph}^{-1} = BT\omega(k)e^{-\frac{C}{T}}$$

- Baseline impurity:

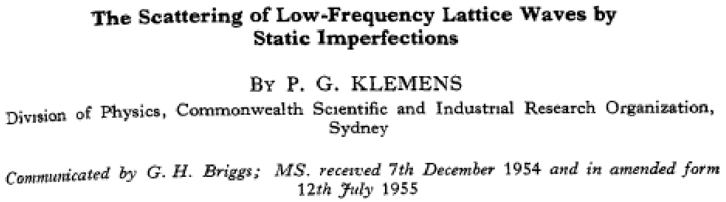
$$\tau_{Imp}^{-1} = A\omega(k)^4$$



Disorder Scattering – Klemens Model

- Phenomenonelogical model developed in 1955, extensively used for thermal conductivity modeling

$$\tau_{Def}^{-1} = \frac{\omega(k)^4 \delta^3 \Gamma_i}{4\pi v_j(k)} \rightarrow Def\ can\ be\ vacancies\ or\ mass - impurities$$


The Scattering of Low-Frequency Lattice Waves by Static Imperfections

By P. G. KLEMENS

Division of Physics, Commonwealth Scientific and Industrial Research Organization, Sydney

Communicated by G. H. Briggs; MS. received 7th December 1954 and in amended form 12th July 1955

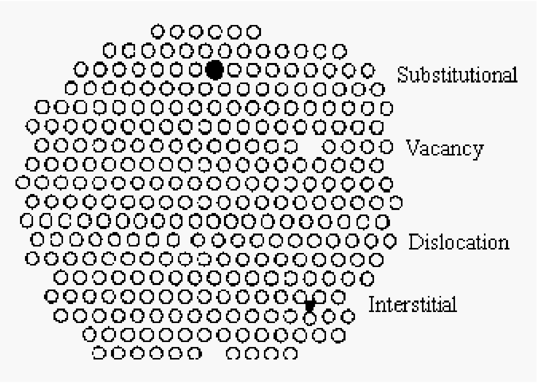
- Scattering cross section of impurity, i:

$$\Gamma_i = x_i \left[\left(\frac{\Delta M_i}{M} \right)^2 + 2 \left(\left(\frac{\Delta G_i}{G} \right) - 2 * 3.2\gamma \left(\frac{\Delta \delta_i}{\delta} \right) \right)^2 \right]$$

  - x_i: defect concentration
  - M: mass of lattice atom
  - G: stiffness constant of nearest neighbor atoms
  - δ: atomic radii

- We expect two kinds of impurities → vacancies, interstitials
- Since we self-irradiated: $\Delta G_i \approx \Delta\delta_i \approx 0$
- For $M_{Vac} = M_{Host}$: $\Gamma_i = x_i[(-3)^2 + 0]$

- *But what is x_i ?? Cannot Predict with TRIM*



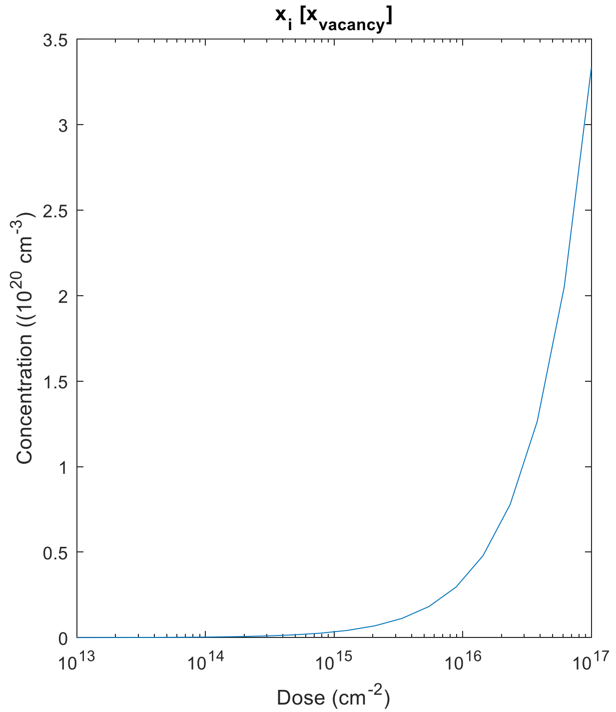
http://chemed.chem.purdue.edu/genchem/topicreview/bp/materials/defects3.html

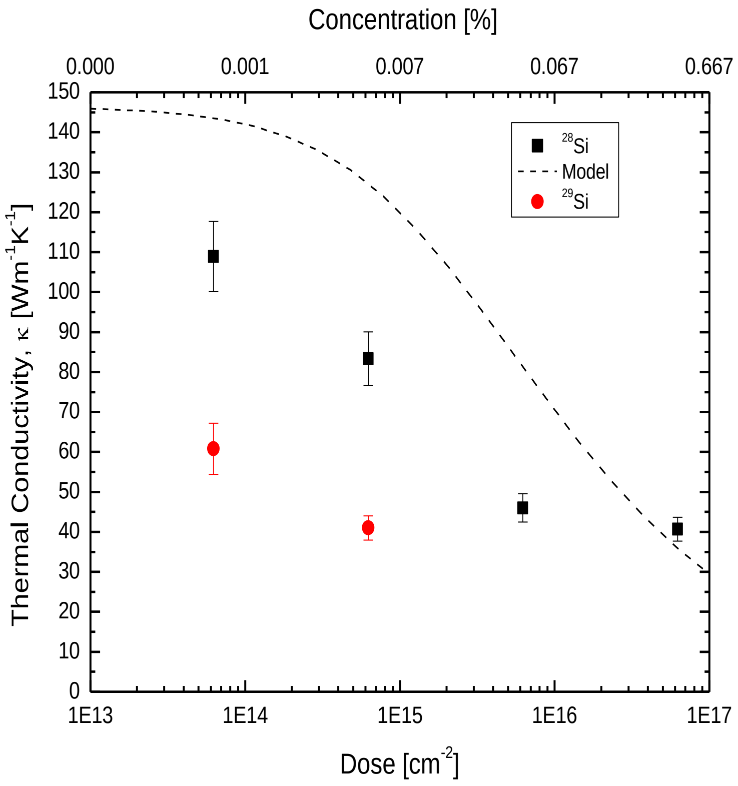
Defect Concentration, x_i

$$\tau_{Def}^{-1} = \frac{\omega(k)^4 \delta^3 \Gamma_i}{4\pi v_j(k)}$$

$$\Gamma_i = x_i \left[\left(\frac{\Delta M_i}{M} \right)^2 + 2 \left(\left(\frac{\Delta G_i}{G} \right) - 2 * 3.2\gamma \left(\frac{\Delta \delta_i}{\delta} \right) \right)^2 \right]$$

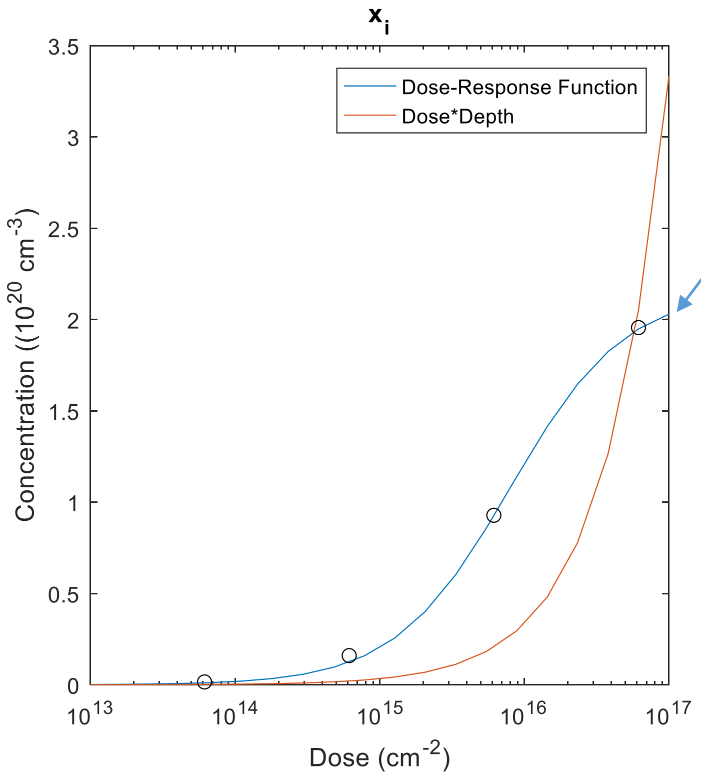
- Ion concentration is known, vacancy concentration is not.
  - If a 1:1 vacancy to ion concentration is assumed:





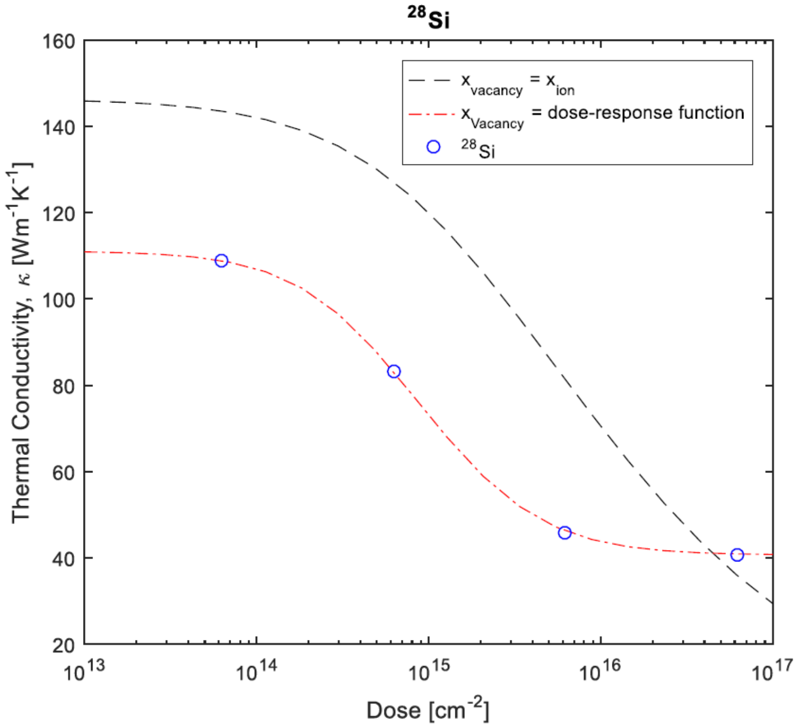
Defect Concentration, x_i

- Instead, calculate x_i for each measured thermal conductivity – then, fit to the calculated values:
  - “Dose Response Function”

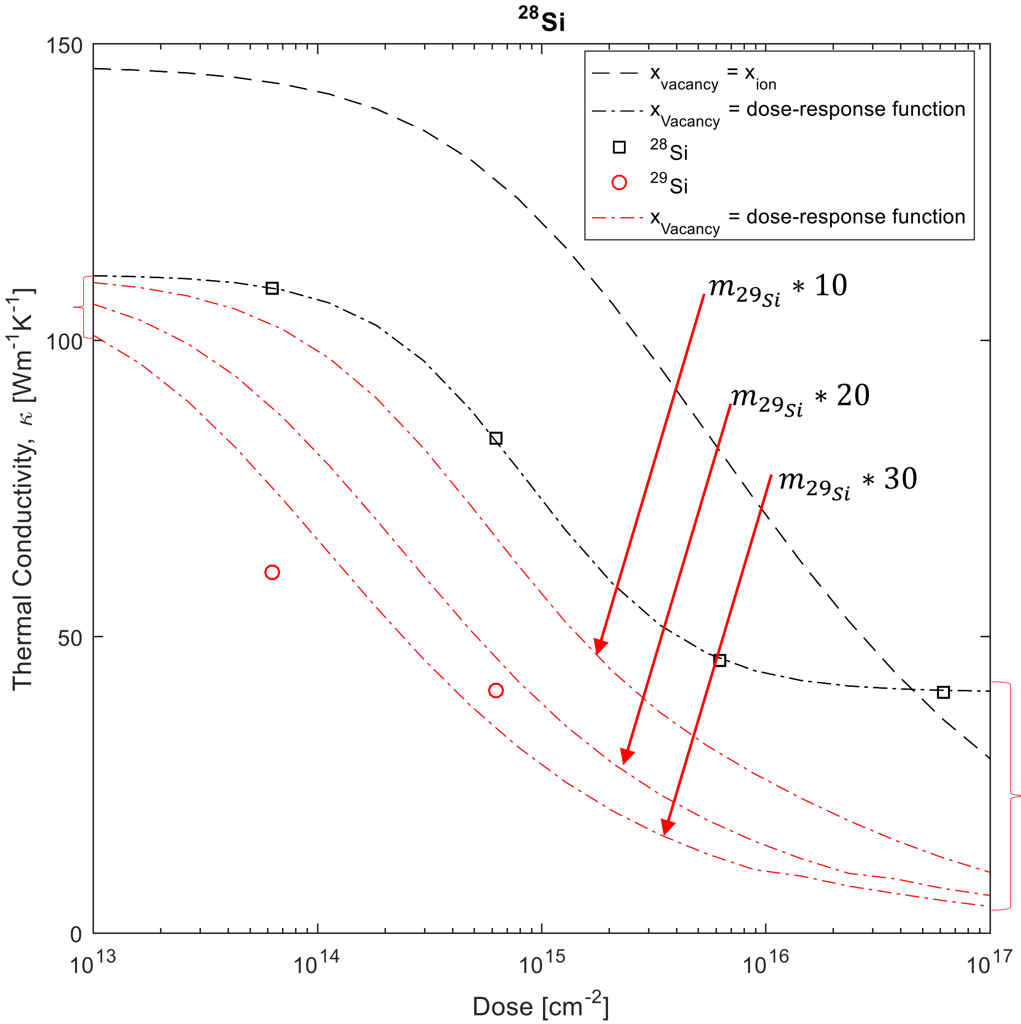


$$x_i = \frac{b-a}{1+\left(\frac{c}{x}\right)^d}$$

- a = min. asymptote
- b = max. asymptote
- c = middle dose value
- d = slope



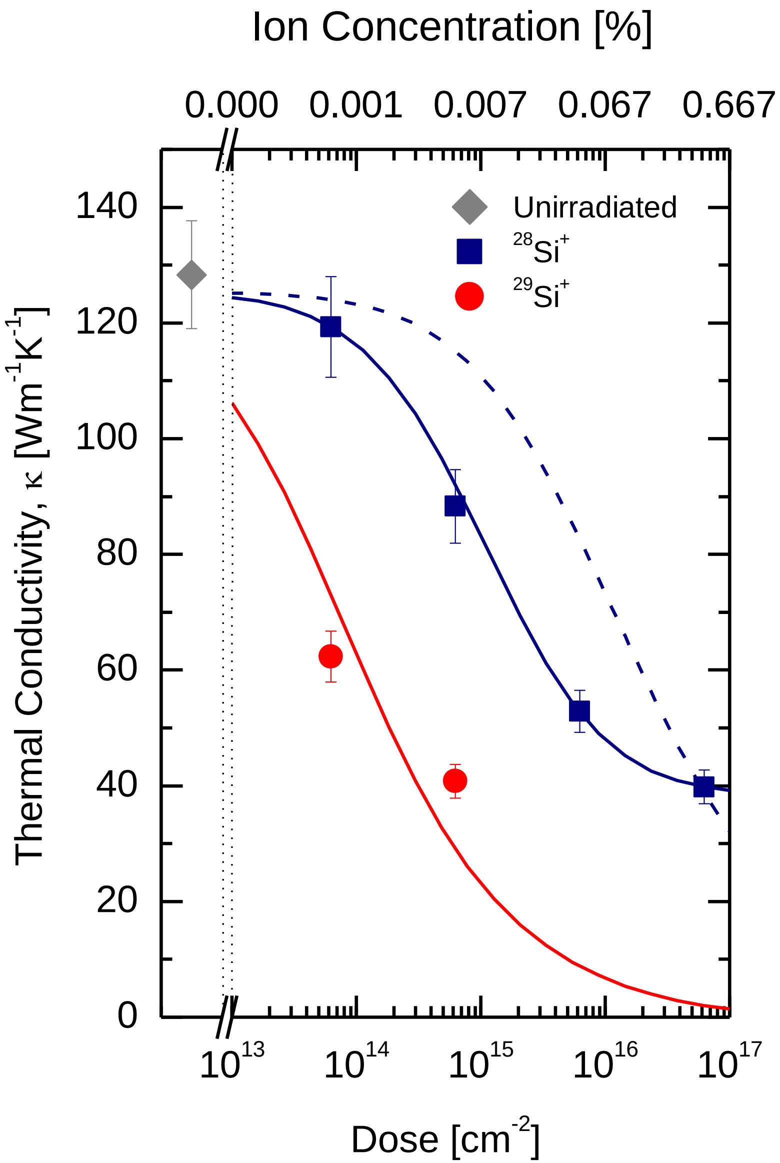
Mass-Impurity Contribution



- Clearly, simply adjusting the mass-impurity term does not capture the trend in the data
- Also consider dislocations and dislocation cores (ω, ω^3)

Assume:

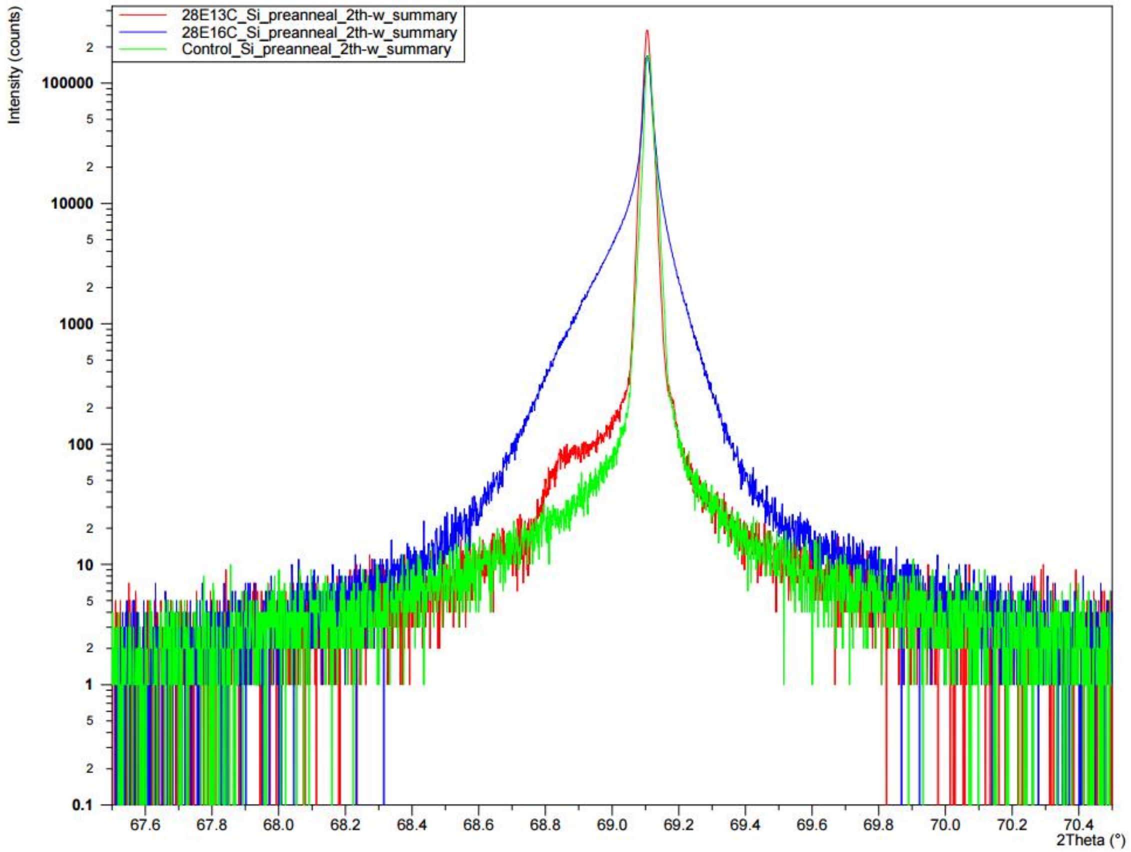
$$\tau_{Def}^{-1} = x_{ion}(A\omega^4 + B\omega^2 + C\omega^3 + D\omega)$$



Strain, Amorphization

- Thermal conductivity model still not within uncertainty of model
  - Disparity due to presence of amorphous regions
  - XRD used to characterize amorphization

- Clear trends are evident with increasing dose
- ^{28}Si:
  - No dose– clean, single peak
  - Low dose (6.24E13) – secondary peak formation indicating strain
  - High dose(6.24E16) – significant peak broadening, indicative of amorphization of sample



Summary / Future Work

- Significant decrease in thermal conductivity when irradiating with heavier isotope

- Decrease cannot solely be attributed to mass differences as predicted by Klemens model – must be a change in vacancy concentration / amorphization
  - Annealing and XRD to yield further insight

- Thermal boundary conductance increases with dose
  - Shown previously in literature (Gorham, et. al) – attributed to spatial gradation of native oxide into Si, leading to region of average vibrational spectra
  - Further testing needed to verify that oxide mixing is responsible for increase