Advanced Non-Destructive Assessment Technology to Determine the Aging of Silicon Containing Materials for Generation IV Nuclear Reactors

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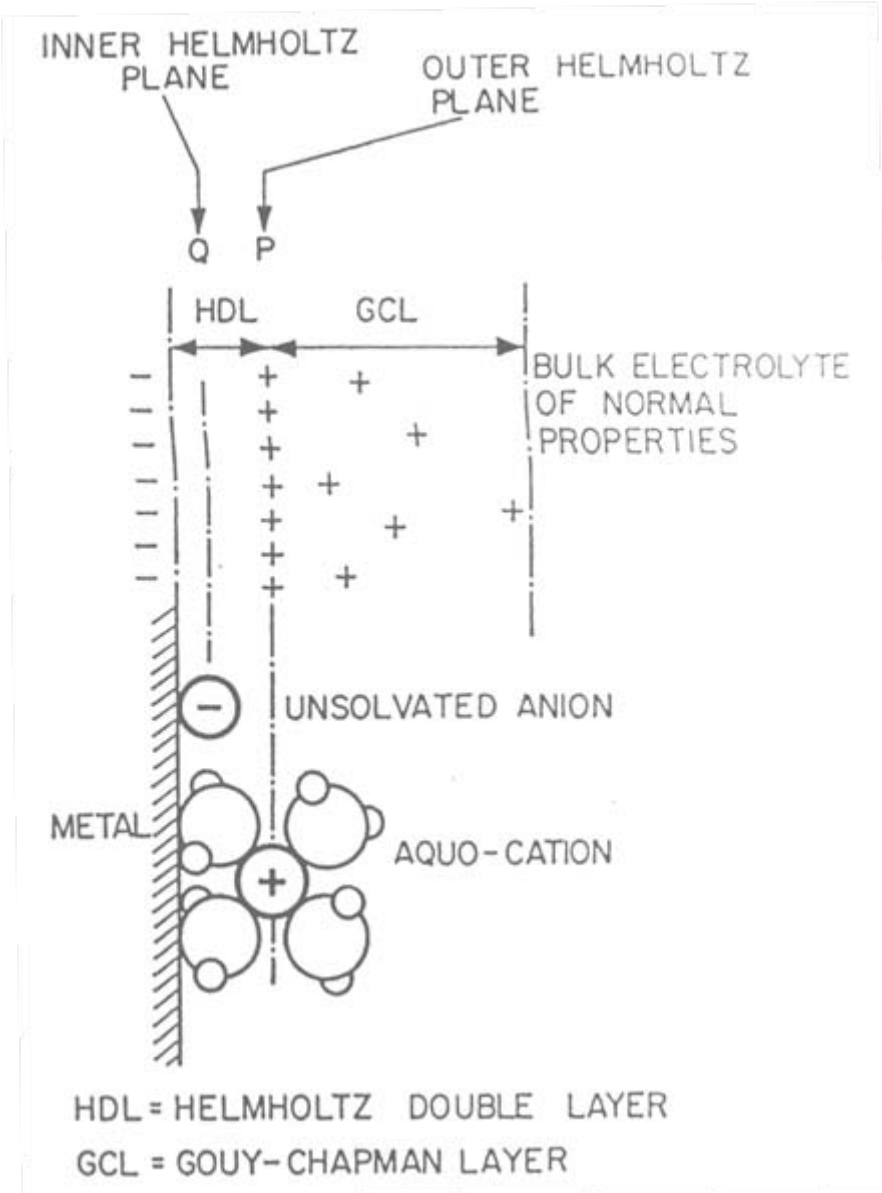
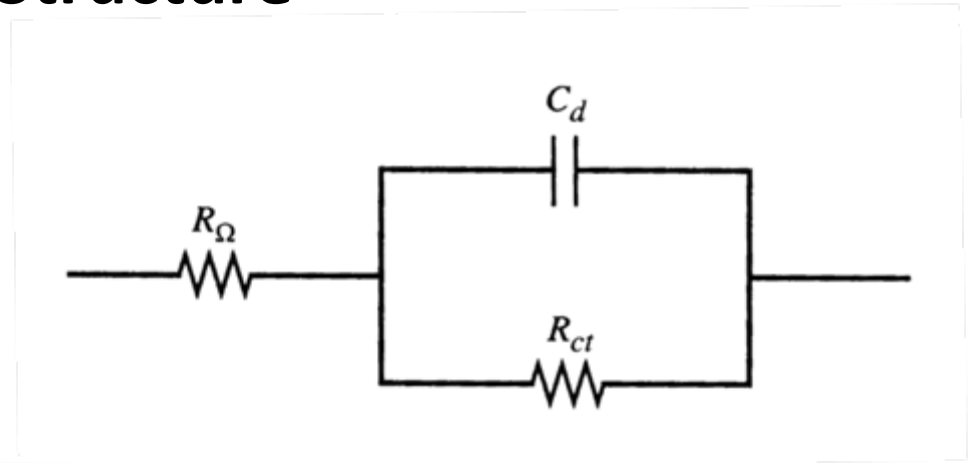
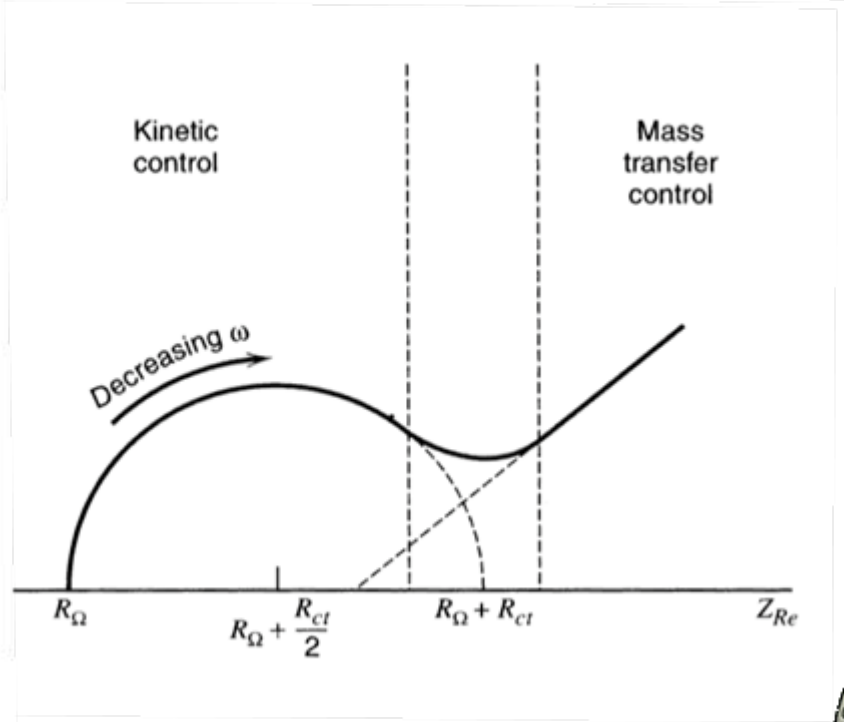
Outline

- Non-linear description of material state awareness
- NDE in-situ detectors for nuclear reactors
- Design criteria
- Silicon carbide as a sensor material
- Effects of irradiation
- Advanced non-destructive measurement techniques
- Results and Analysis
- Conclusions

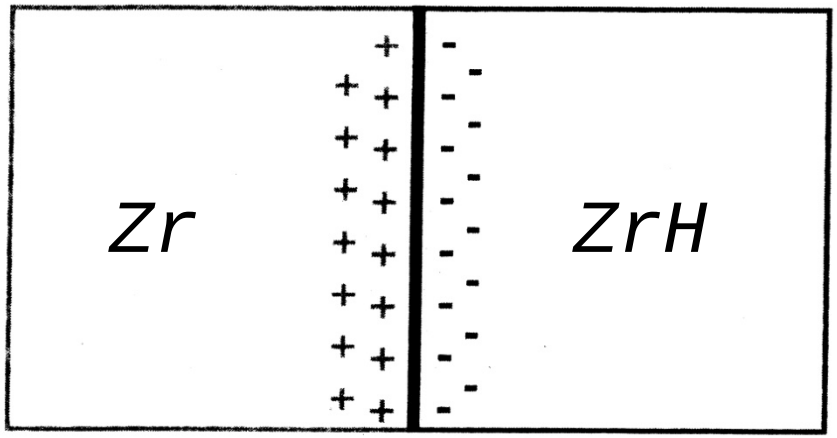
Electrochemical Impedance Spectroscopy: Double Layer Structure





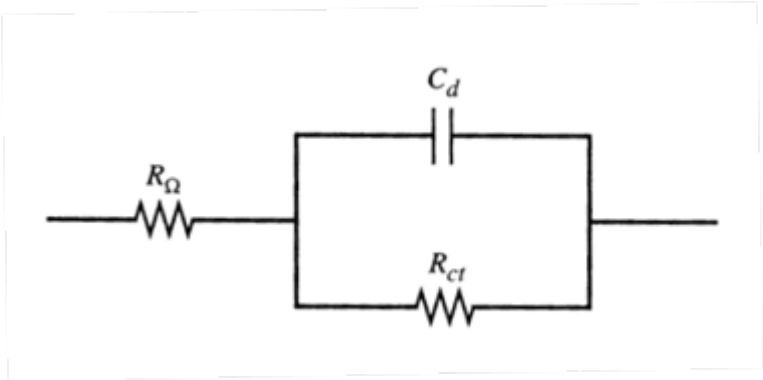


Microstructural Junctions

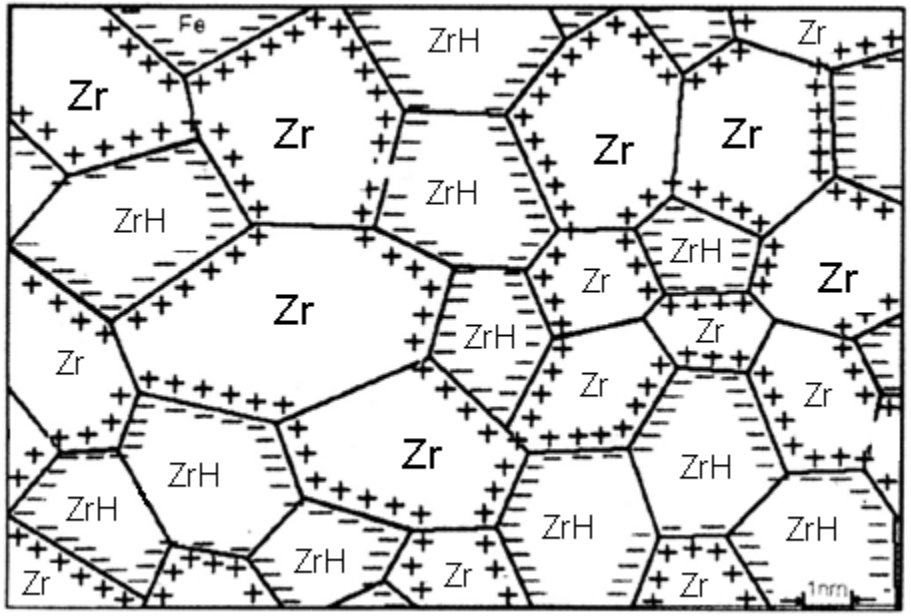


- *Thermodynamic equilibrium dictates that the electrochemical potential be the same on both sides of the interface.*
- *The electrochemical potential difference is balanced by an electrostatic potential*
- *Space charge layer is on the order of 0.5 nm*

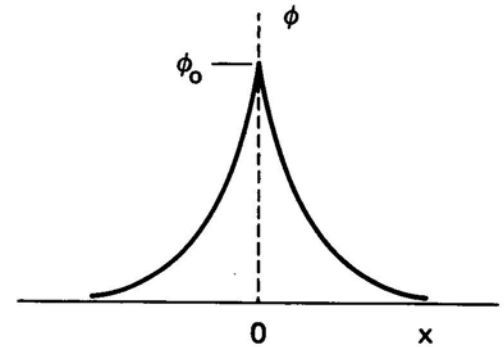
Could microstructural junctions be modeled as an RC circuit ?



Adopted from: Gleiter, 2009



Poisson's Equation



- Consider the e^- distribution associated with a defect in the lattice

$$\varepsilon_0 \frac{d^2\phi}{dx^2} = \rho e n_\infty \{[\exp(\frac{e\phi_0}{kT})] - 1\}$$

- Poisson's Equation

$$\frac{e\phi_0}{kT} << 1$$

Apply Taylor Serie

$$\varepsilon_0 \frac{d^2\phi}{dx^2} = \rho e n_\infty [(\frac{e\phi_0}{kT}) + \frac{1}{2}(\frac{e\phi_0}{kT})^2 + \ldots]$$

$$\varepsilon_0 \frac{d^2\phi}{dx^2} = \frac{\rho e^2 n_\infty \phi_0}{kT}$$

- 1st term solution

$$\phi = \phi_0 \exp\frac{-|x|}{\lambda_o}$$

Ref: Tu, 1985; Chen, 1983

Below λ_o need 2nd order term

Debye length

$$\lambda_o = (\frac{\varepsilon_0 kT}{\rho e^2 n_\infty})^{\frac{1}{2}}$$

Dislocation Potential Field

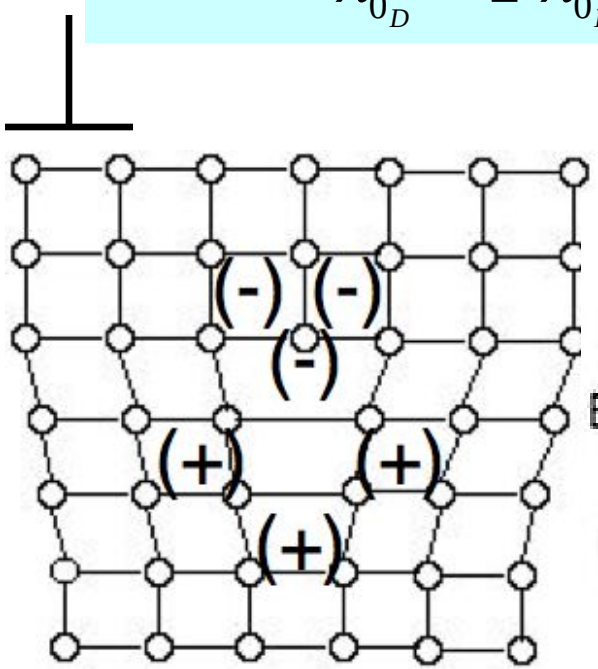
How does dislocation interact with electric field?

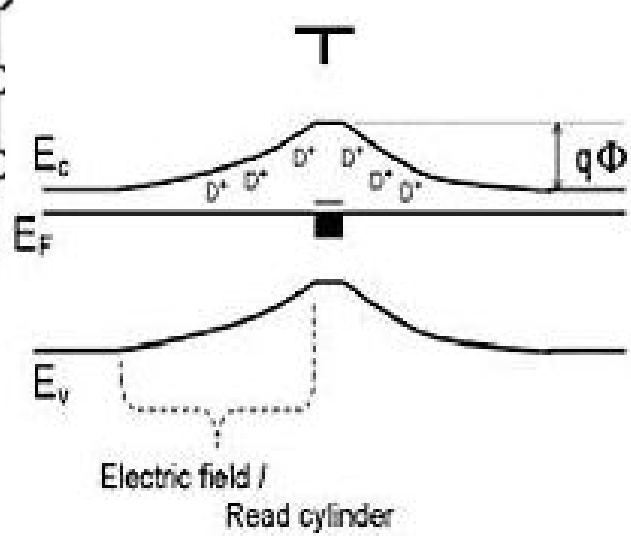
$$\phi = \phi_0 \exp\frac{-|x|}{\lambda_o} \longrightarrow \phi = \phi_0\left(\frac{x}{\lambda_0} - \frac{1}{2}\frac{x^2}{\lambda_0^2} + \frac{1}{6}\frac{x^3}{\lambda_0^3} - \ldots\right)$$

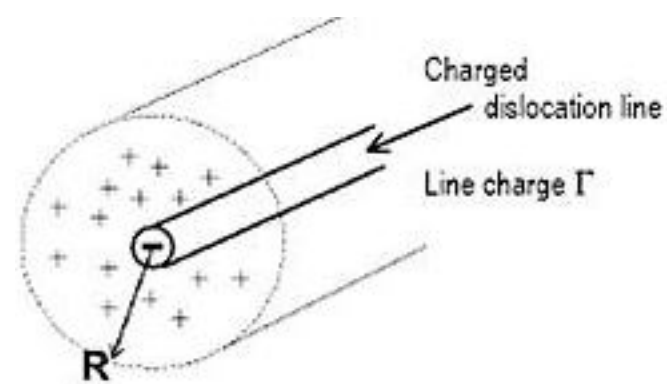
$$\phi_D = \phi_{0_D}\left(\frac{x}{\lambda_{0_D}} - \frac{1}{2}\frac{x^2}{\lambda_{0_D}^2} + \frac{1}{6}\frac{x^3}{\lambda_{0_D}^3} - \ldots\right)$$

$$\lambda_{0_D} = \left(\frac{\varepsilon_0 kT}{\rho_D e^2 n_\infty}\right)^{\frac{1}{2}}$$

$$F_D = -\frac{d\phi_D}{dx}$$





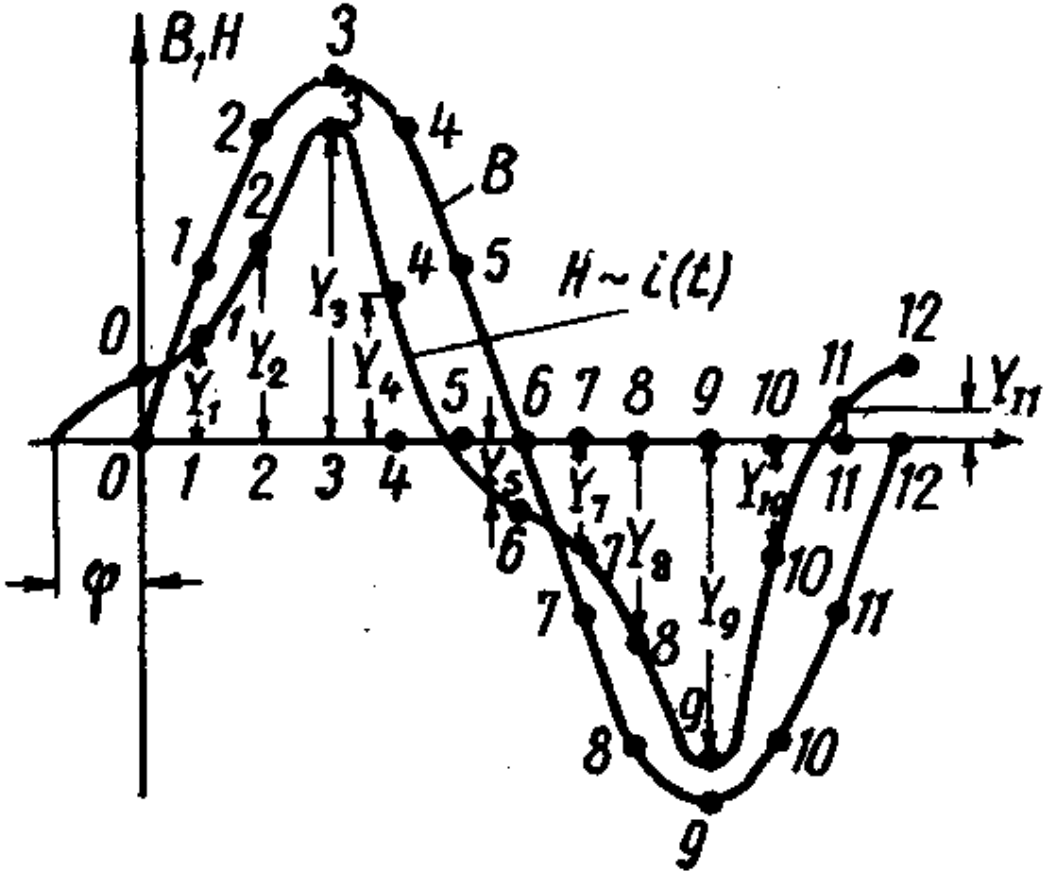


Wave Analysis - FM, AM, and Harmonics

- ***Frequency Modulation*** $\nu\lambda = c$
  - *Wavelengths - Nano, micro, meso microstructural features assessments*

- ***Amplitude Modulation*** $V = V_o \sin \omega t$ $\varepsilon = \varepsilon_o \sin \omega t$
  - *V_o variation,* ε_o *Variation*
  - *Relaxation -> V = exp (-kt),* $\varepsilon_o = \exp(-kt)$
  - ***Can determine kinematic behavior (diffusion coefficient, dynamic recovery)***

Harmonic Analysis of Induced Voltage



$$i(t) = \sum_{k}^{\infty} (A_k \sin k\omega t + B_k \cos k\omega t)$$

Gruska 1983

Nonlinear Wave Analysis

Consider the nonlinear wave equation:

$$\frac{\partial^2 u}{\partial t^2} = c^2 \frac{\partial^2 u}{\partial x^2} - \beta c^2 \frac{\partial u}{\partial x}\frac{\partial^2 u}{\partial x^2}$$

Assume a solution of the form:

$$u(x,t) = \sum_{k=0}^{\infty} \beta^k u_k(x,t) = u_0(x,t) + \beta u_1(x,t) + \beta^2 u_2(x,t) + \mathsf{L}$$

For this example, take only the first two terms

$$u(x,t) = u_0(x,t) + \beta u_1(x,t)$$

Second harmonic generation

Solution to the linear Wave Equation

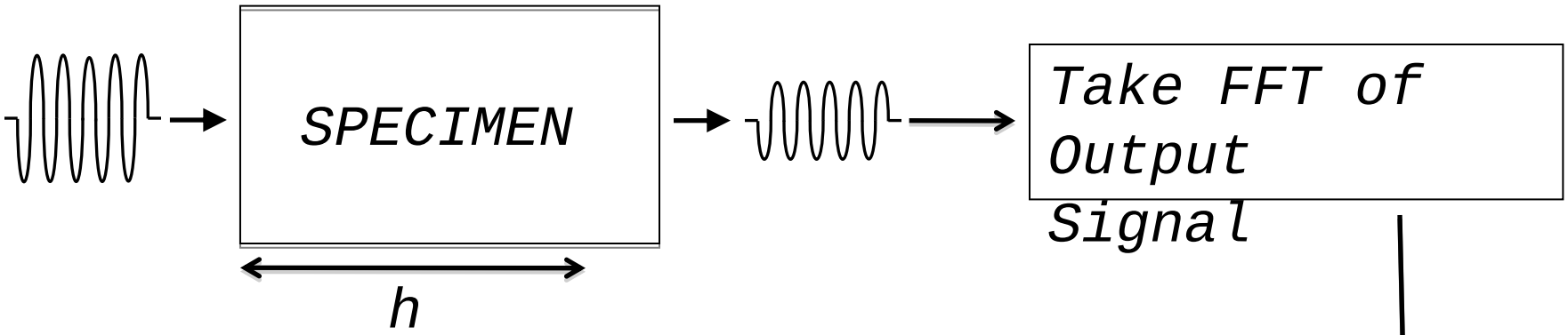
Assuming a solution of the form: $u_0(x=0,t) = p_0 \cos(\omega t)$

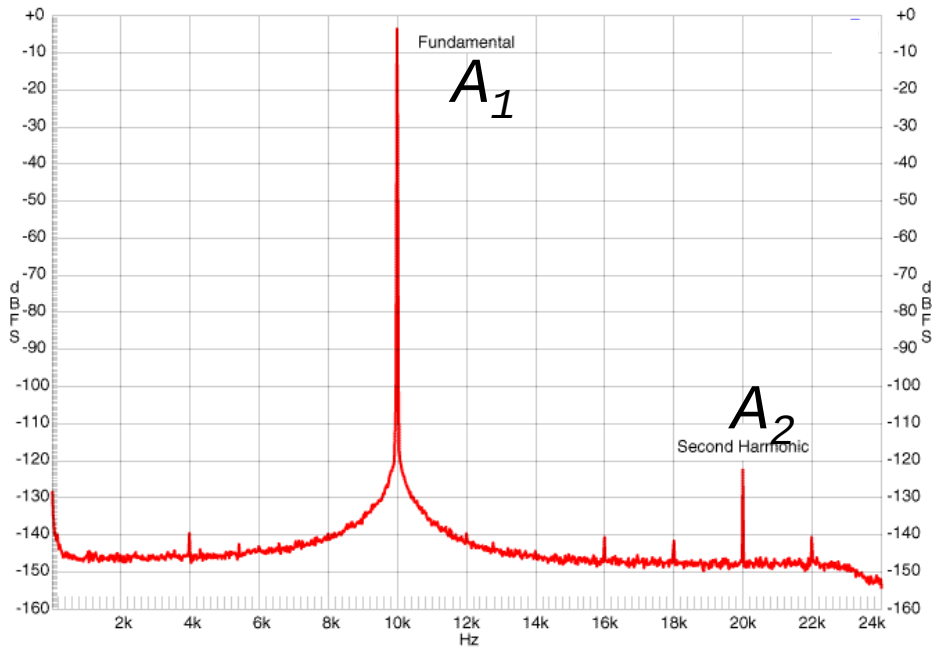
And substituting the series expansion into the governing equation, a final solution is obtained

$$u(x,t) = \frac{1}{8}\beta k^2 p_0^2 x + p_0 \cos(kx - \omega t) - \frac{1}{8}\beta k^2 u_0^2 x \cos(2[kx - \omega t])$$

Second harmonic

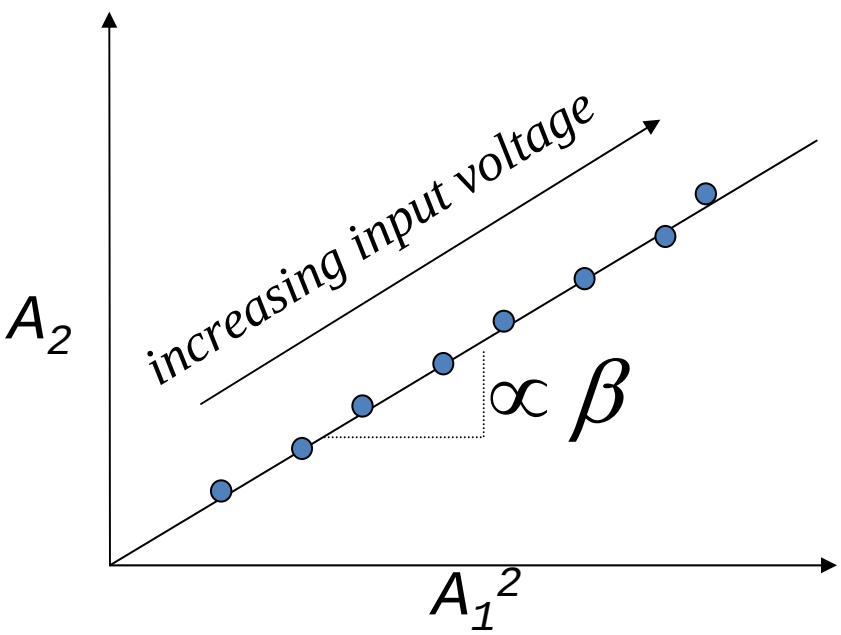
Nonlinearity & Ultrasonic NDE





Nonlinearity Parameter

Determination of the nonlinearity parameter:



$$\beta = \left[\frac{2\,c_l^2}{h\,\pi\,f^2}\right]\frac{A_2(h)}{A_1^2}$$

c_l : longitudinal wave speed

f : frequency

h : Propagation distance (specimen thickness)

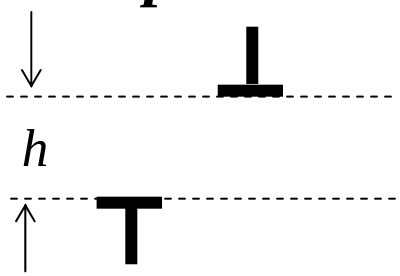
A_1 : *Absolute amplitude* of the fundamental

A_2 : *Absolute amplitude* of the second harmonic

Cantrell,

Dislocations: Dipoles and Precipitates

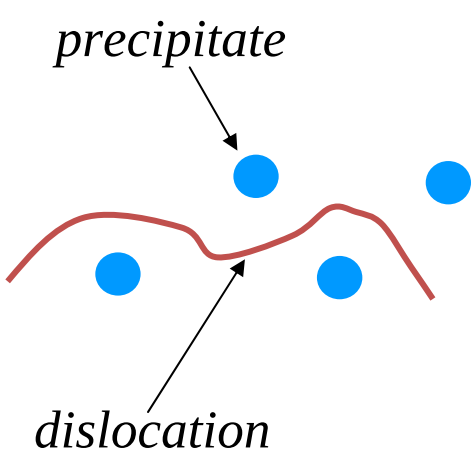
Dipoles



$$\beta_d = \frac{16\pi^2 \rho_d \Omega \left(1-\nu\right)^2 C_{11}^2}{G^2 b}$$

ρ_d = *dislocation (dipole) density*
h = *dipole height*

Precipitates



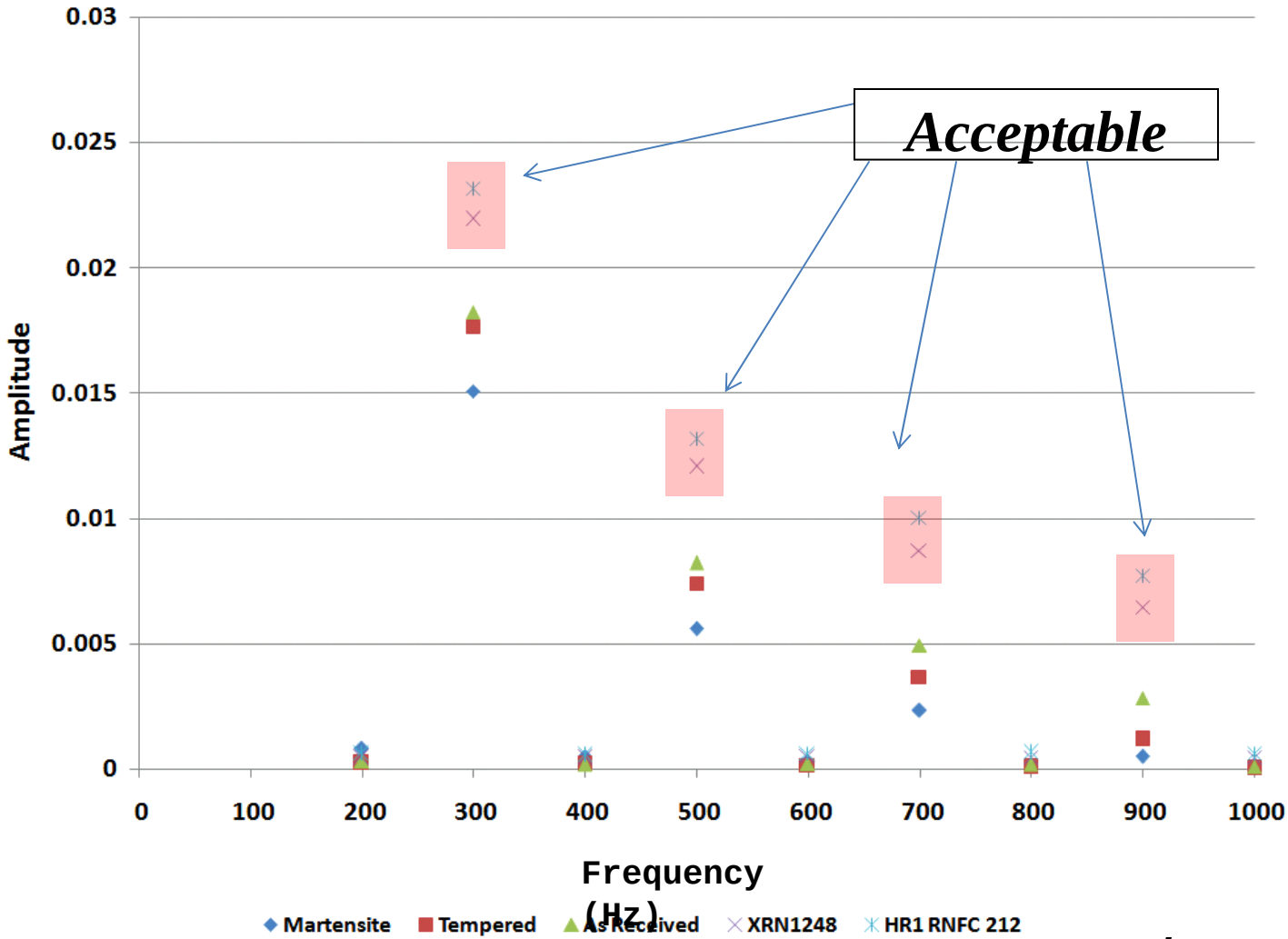
$$\beta_p = \frac{205\, \rho_m\, r^4 \Omega^2 \left(1+\nu\right) C_{11} \left|\delta\right|}{G^2 b^2 \left(1-\nu\right) f_p^{1/3}}$$

r = *radius of precipitates*
δ = *lattice misfit parameter between the precipitate and the matrix (coherency)*
ν = *Poisson's ratio*
f_p = *volume fraction of precipitates*

Cantrell,

Frequency Analysis

FFT of hysteresis measurements – small sensor at a 100Hz.



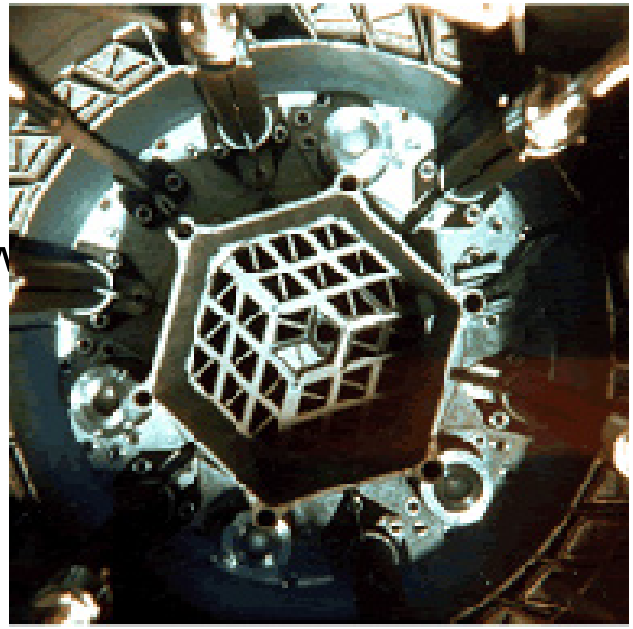
Meir, 2009

QNDE of Reactor Core Materials

- High levels of neutron fluence radiation damage can cause significant degradation to reactor components
- In-situ non-destructive monitoring of reactor components will increase integrity and operating capacity
- Use of QNDE with advanced wave analysis to achieve material state awareness of reactor core materials

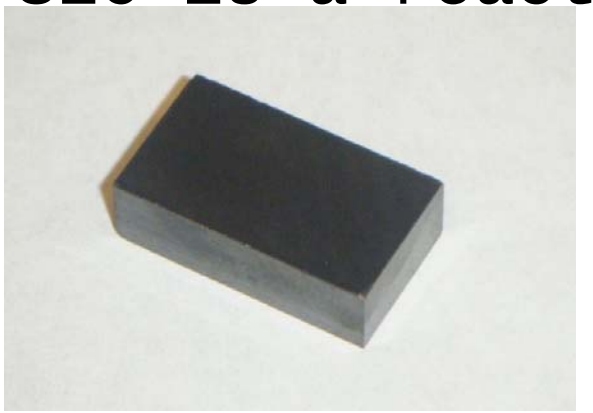
Radiation Damage Monitor Design Criteria

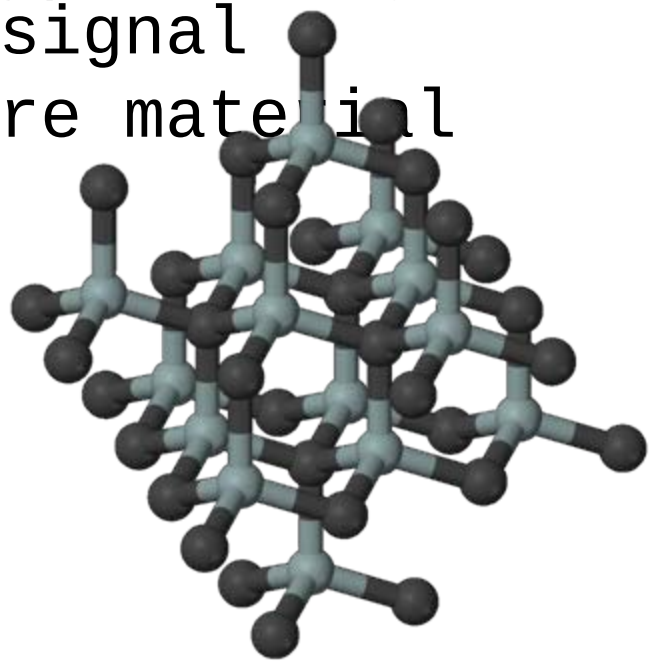
- Small
- Cost Effective
- High Temperature Capability
- Reliable Property Correlation to Fluence
- Replaceable
- Disposable
  - Relatively low activation and half life



Silicon Carbide Sensor Material

- High temperature capabilities
- Neutron doping results in phosphorous formation – semiconductor properties
- Alteration of polytypes can give irradiation damage signal
- SiC is a reactor core material





Effect of Irradiation: Neutronics

- Phosphorus doping achieved through reaction:

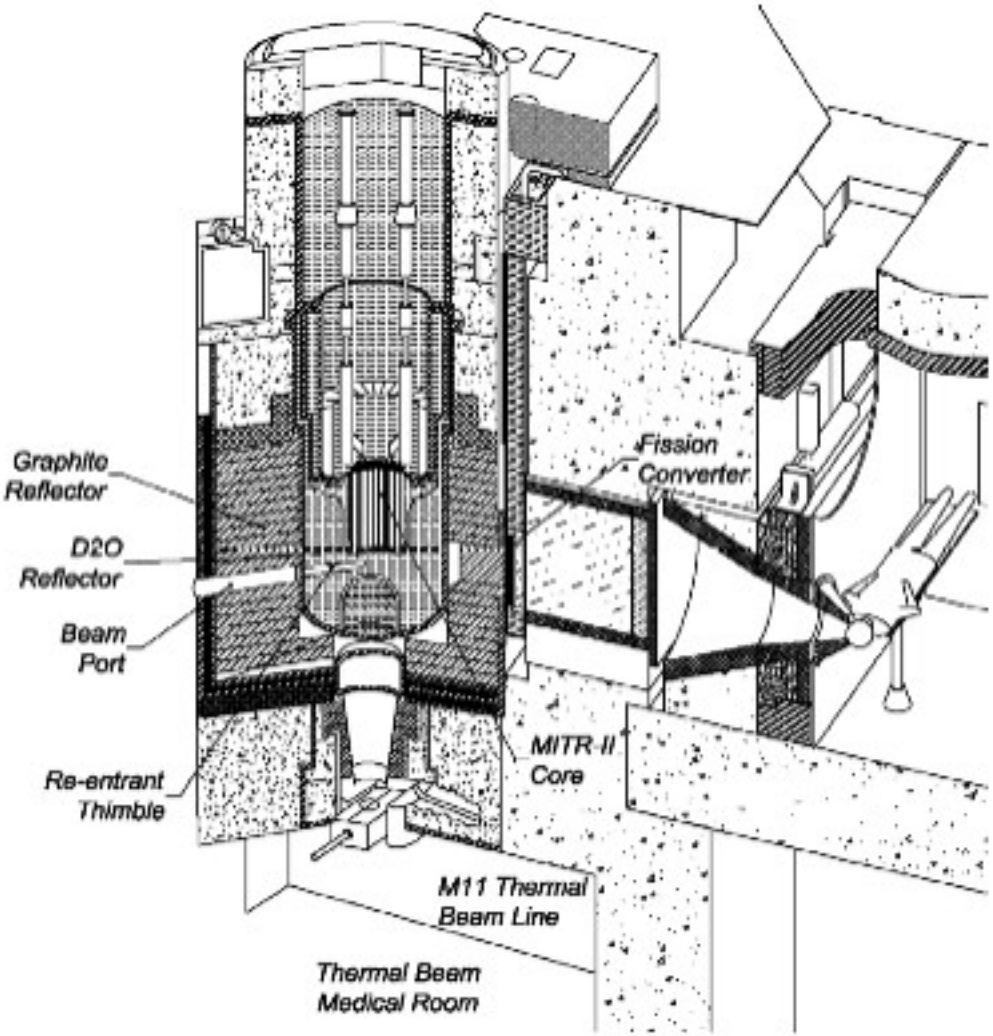
$$^{30}Si + n \rightarrow {}^{31}Si + \gamma \rightarrow {}^{31}P + \beta$$

- Process driven mainly by thermal neutrons

- Overall doping rate dependent on ^{31}Si half life, and rate of P to Si reaction:

$$^{31}P \rightarrow {}^{32}P \rightarrow {}^{32}Si$$

Irradiation Facility:
MIT Reactor

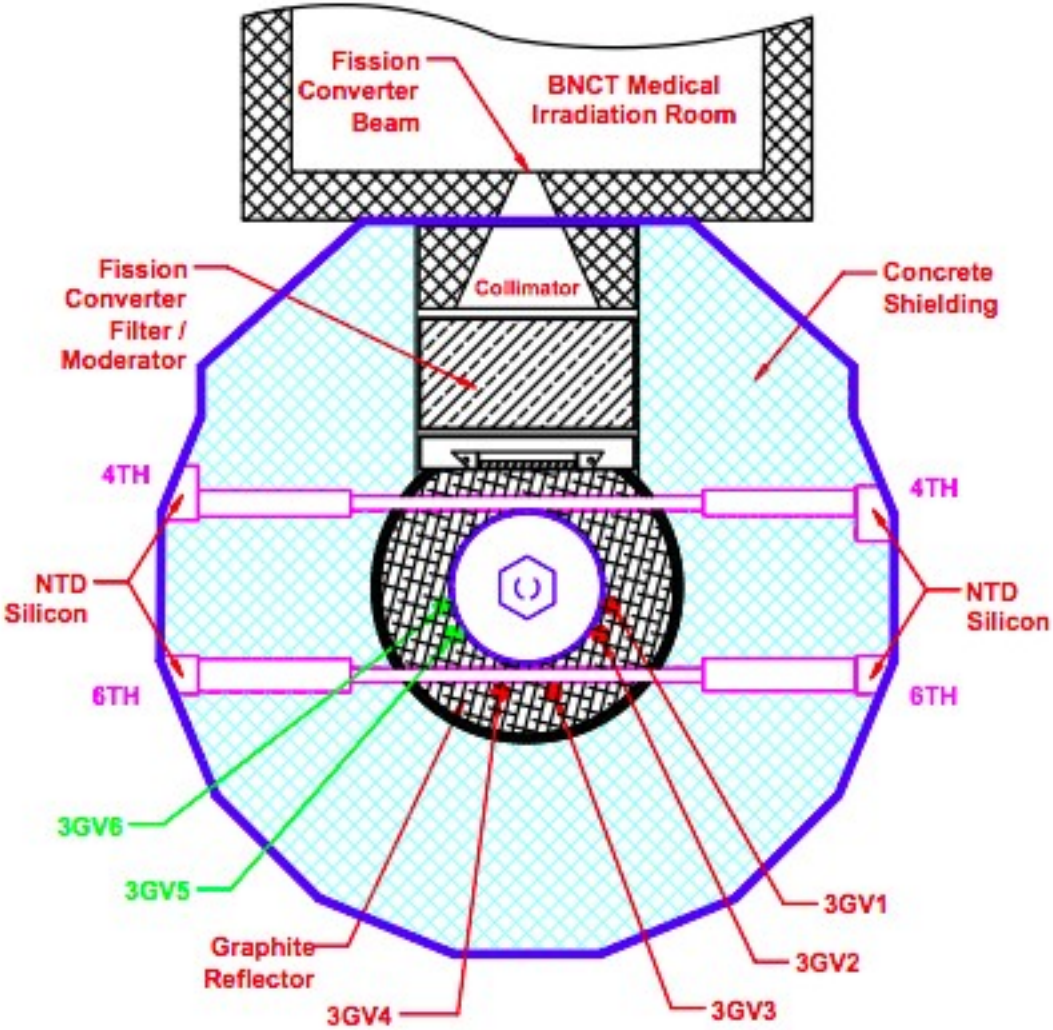


Irradiation Environments

- Sample 1:
  - 3GV6 facility
  - 9.63E+17 neutrons/cm^2
  - Cd ratio of about 200
  - 24 hour irradiation period

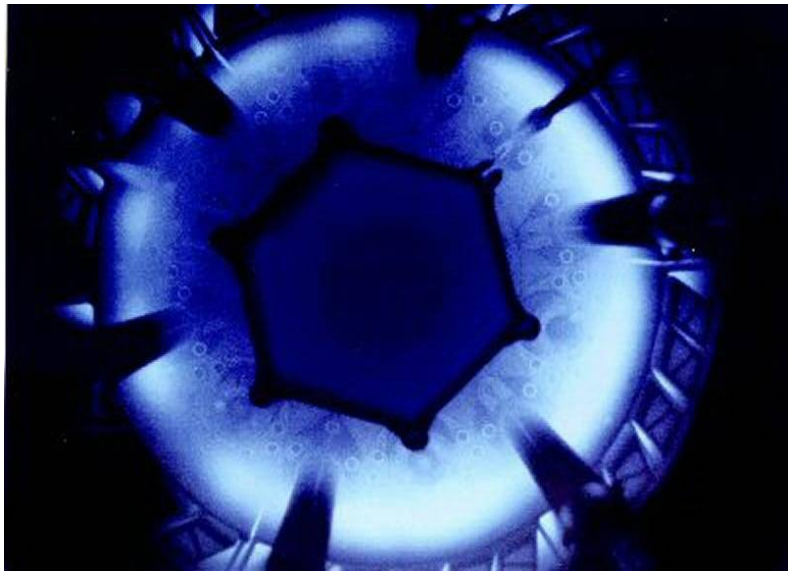
- Sample 2:
  - 3GV6 facility
  - 9.26E+18 neutrons/cm^2
  - Cd ratio of about 200
  - 244 hour irradiation period

Irradiation Environments, cont.



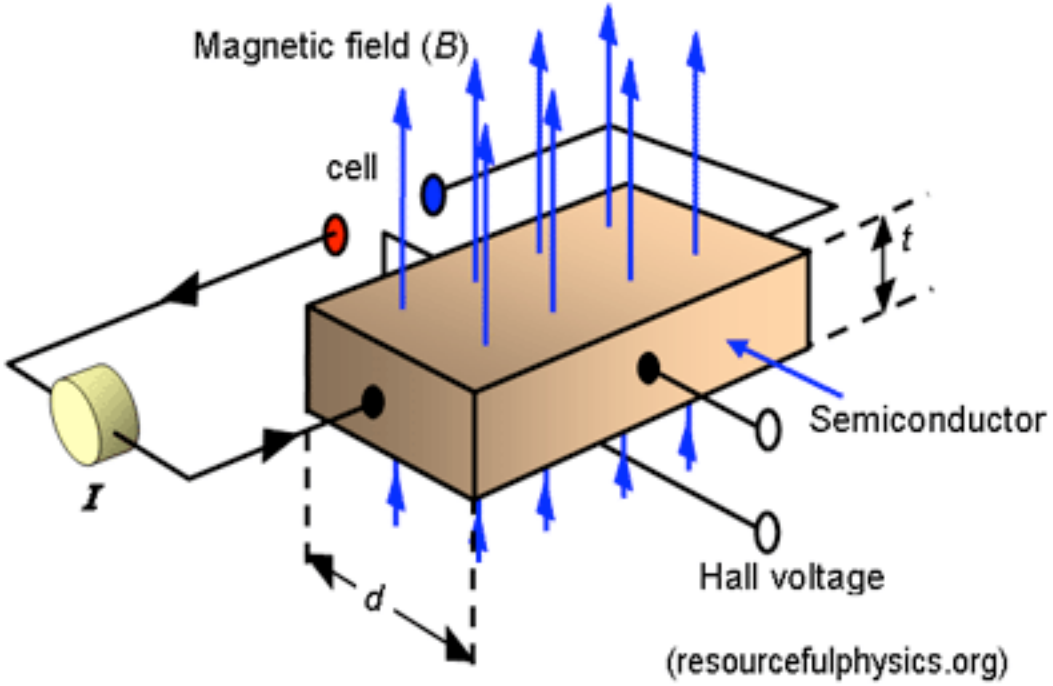
Irradiation Environments, cont.

- Sample 3:
  - 2PH1 facility
    - 5-10X higher flux than 3GV6 facility
  - 2.82E+19 neutrons/cm^2
  - 226 hour irradiation period



Hall Effect

- Hall Effect - establishment of voltage difference in direction transverse to electric current in sample and magnetic field perpendicular to current flow



Hall Effect

- Hall coefficient - ratio of electric field to product of current density and magnetic field:

$$R_H = \frac{E}{jB}$$

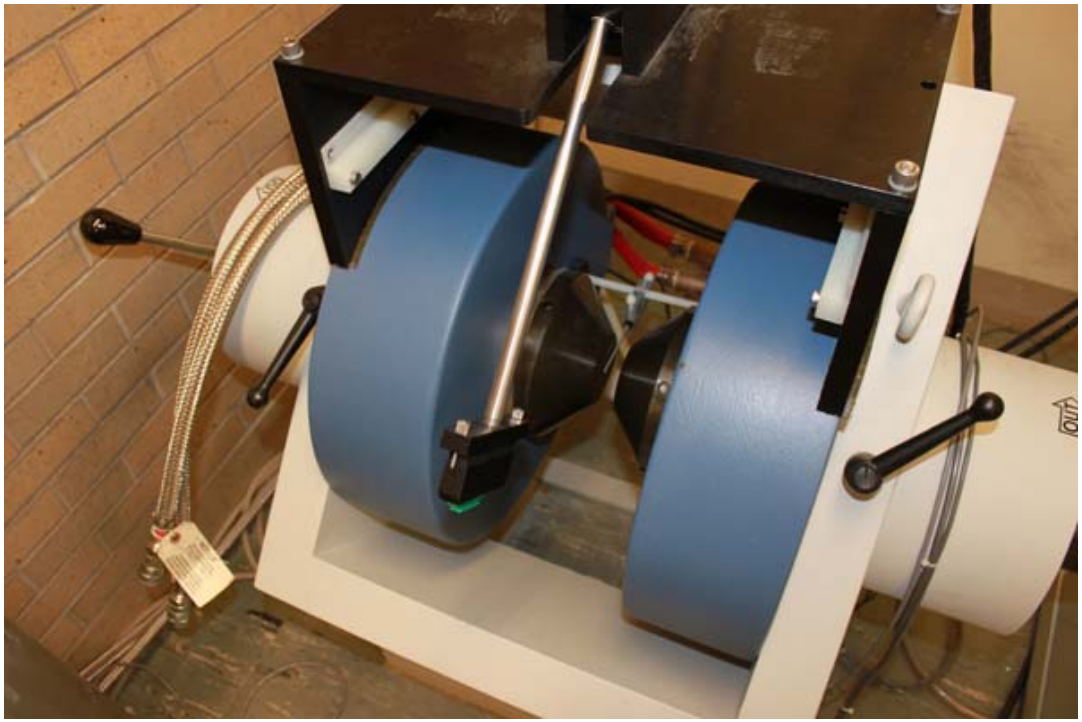
- Hall coefficient complicated by two types of charge carriers seen in semiconductors (holes/electrons):

$$R_H = \frac{-n\mu_e^2 + p\mu_h^2}{e(n\mu_e + p\mu_h)^2}$$

Hall Effect

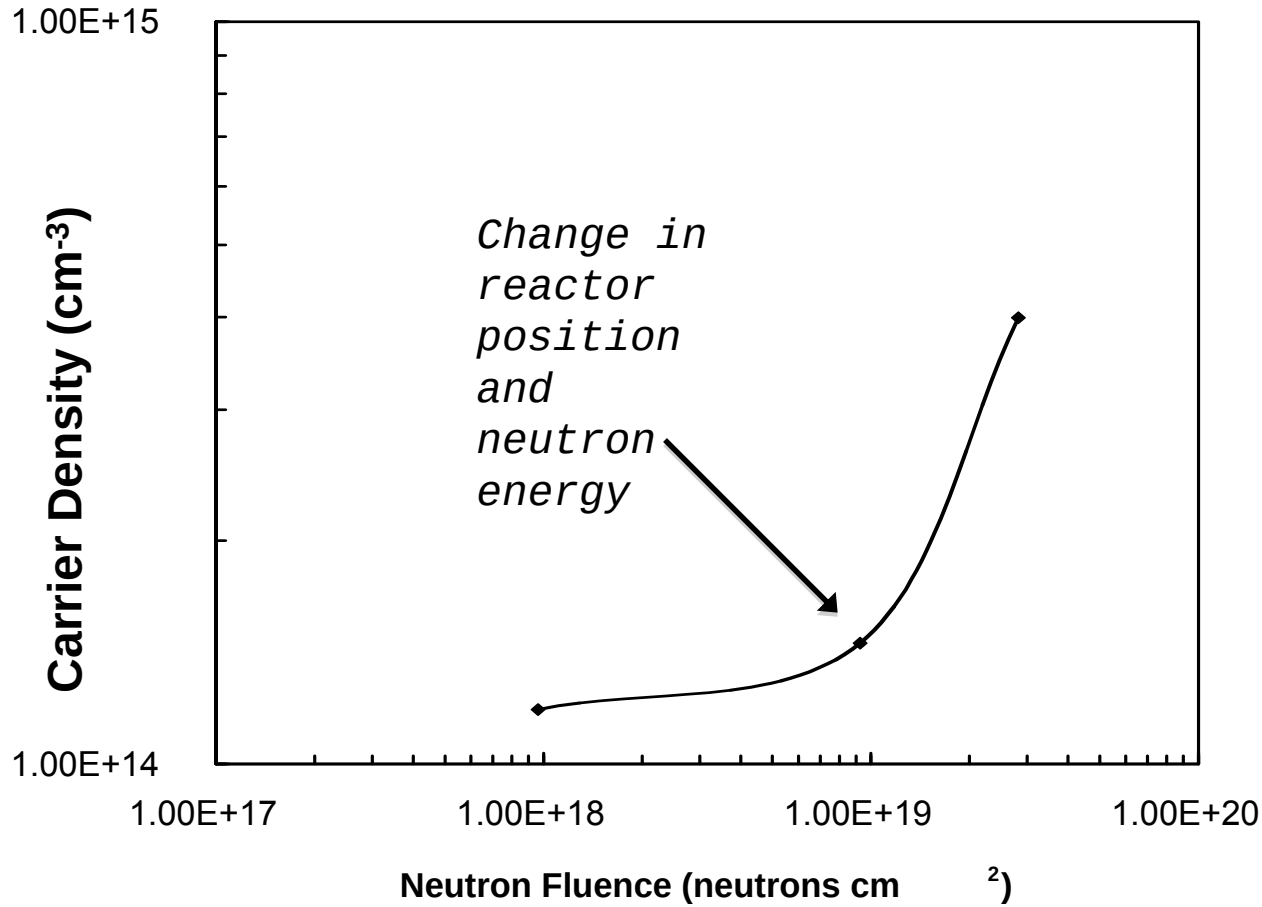
Hall Coefficient Measurements

CSM Physics Department



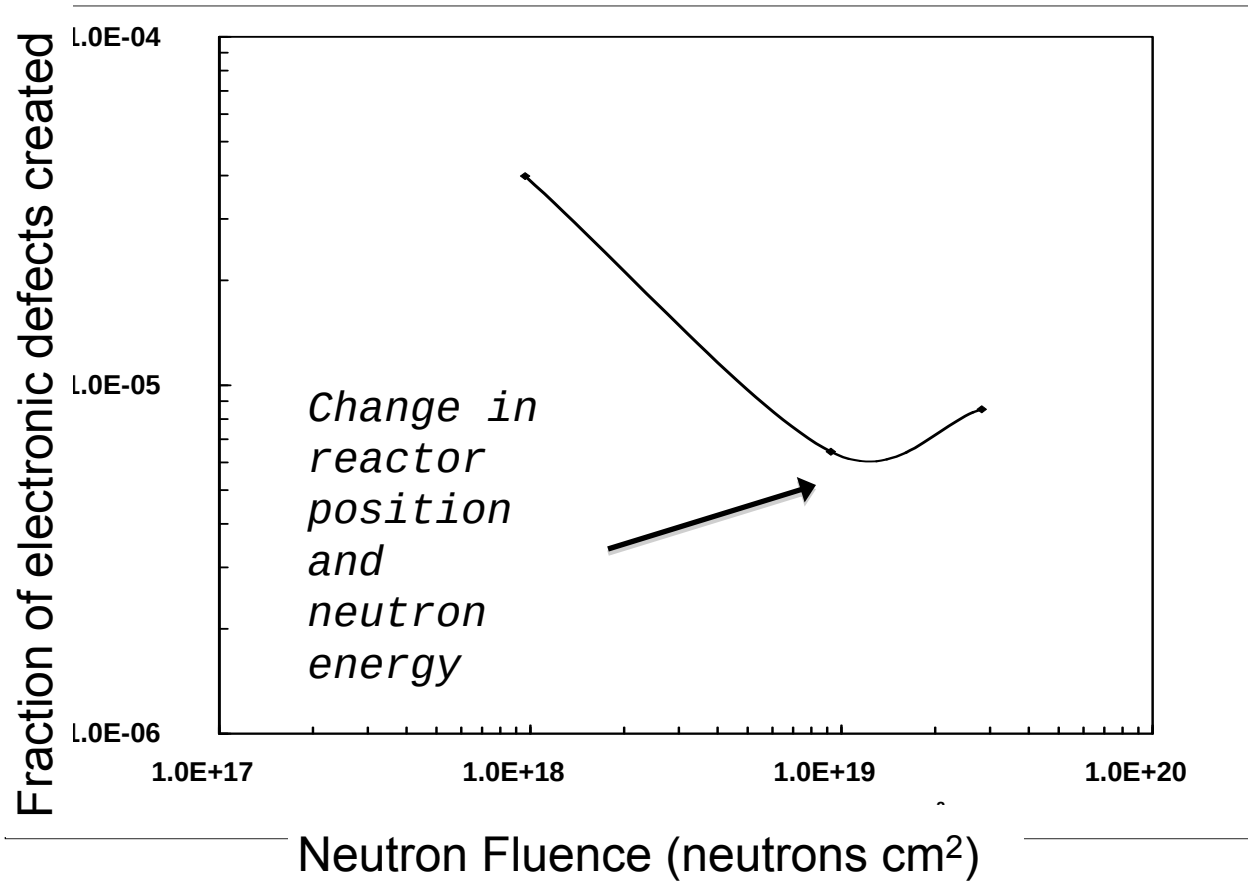
Results and Analysis

Carrier Density Increase



Results and Analysis

Collision Efficiency Decrease



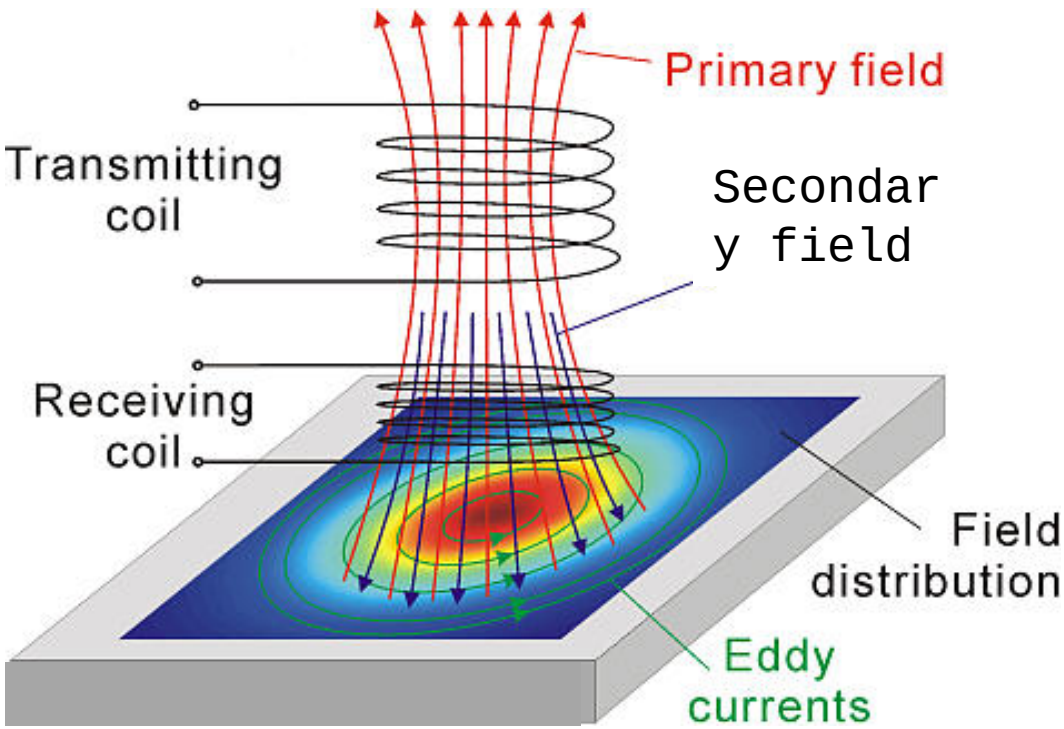
High Frequency Impedance Measurements

$$Z = \sqrt{R^2 + (1/\omega C)^2 + (\omega L)^2}$$

Resistance

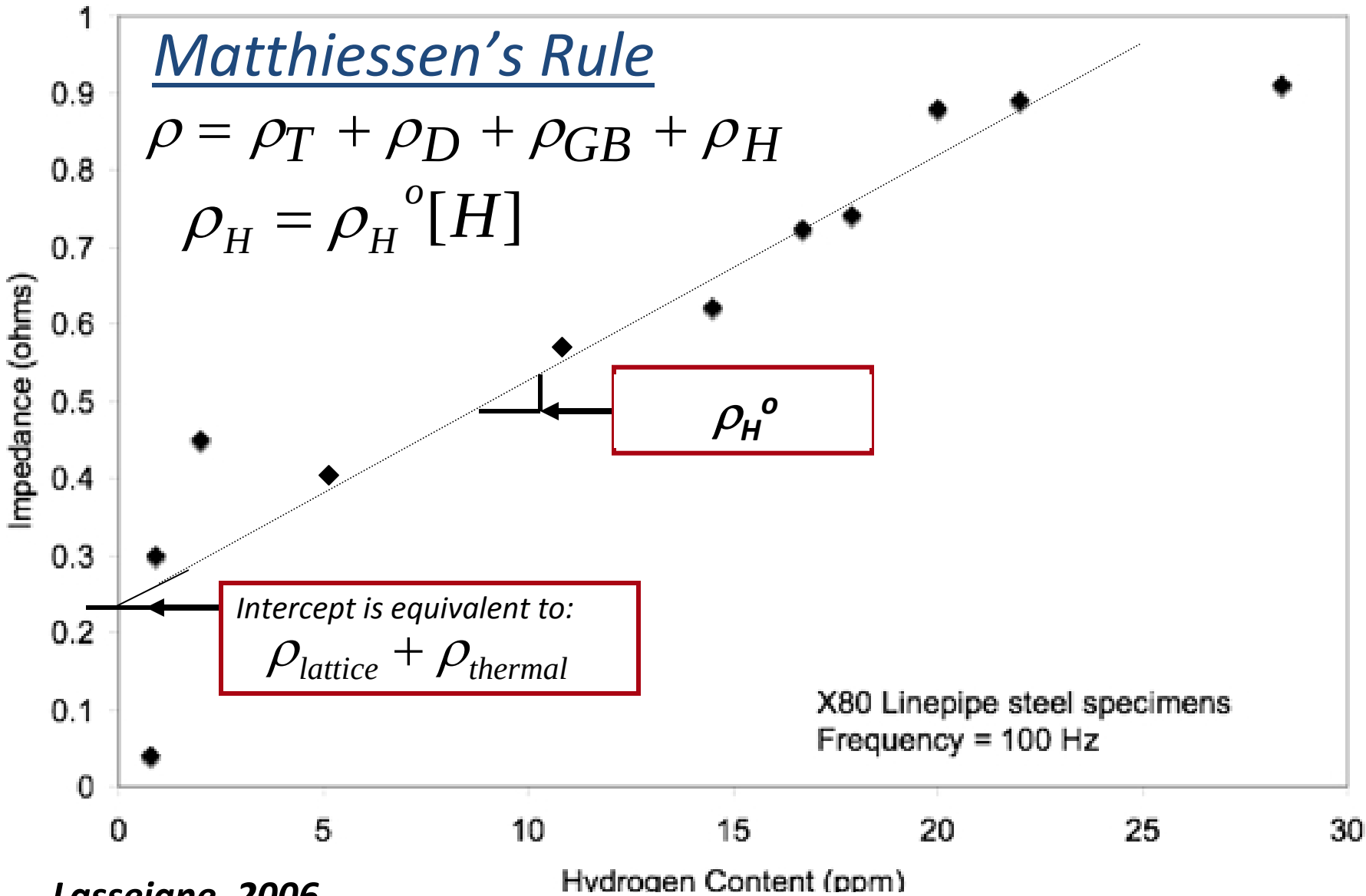
Capacitance Reactance

Inductance Reactance



B.P.C. Rao, 2006

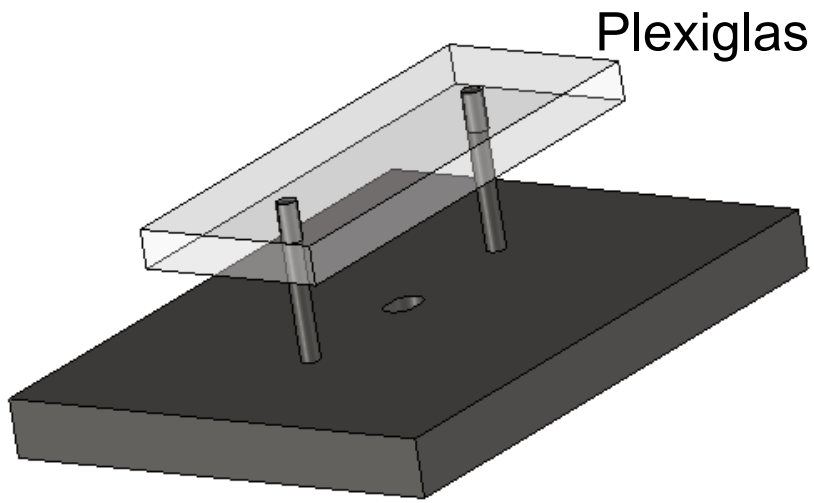
Impedance as a Function of Hydrogen



Lasseigne, 2006

High Frequency Impedance Analysis Technique

- Sample holder created to ensure testing consistency
  - AISI A-36 steel base, Plexiglas for suspension of pancake coil

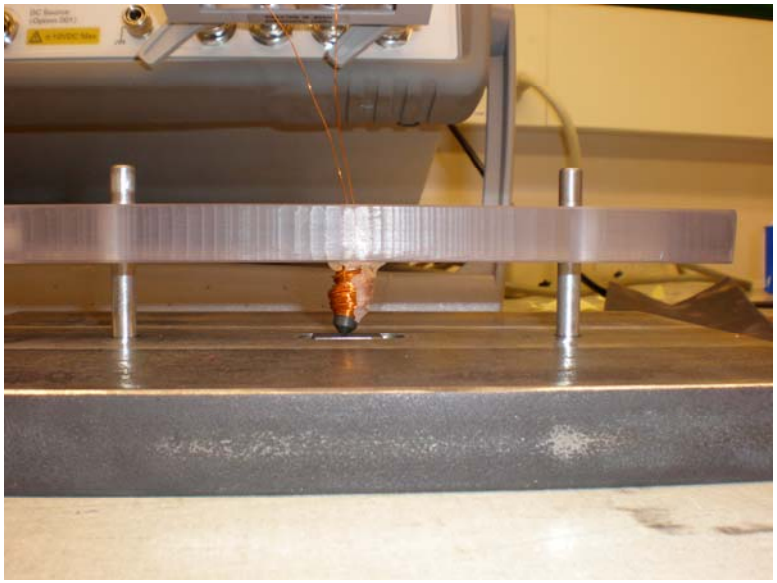


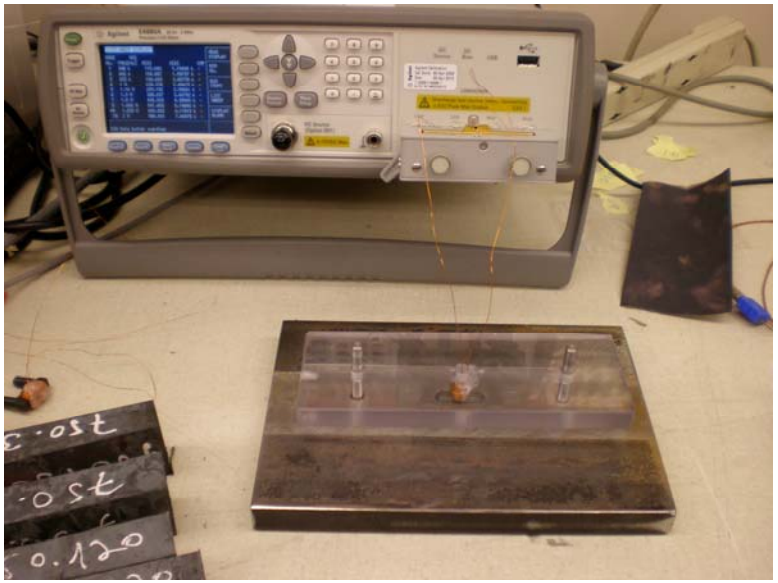
$$D = [1/(f\sigma\rho)]^{1/2}$$

Steel base was needed since the impedance reflection depth of SiC is ~11 m

High Frequency Impedance Analysis Technique

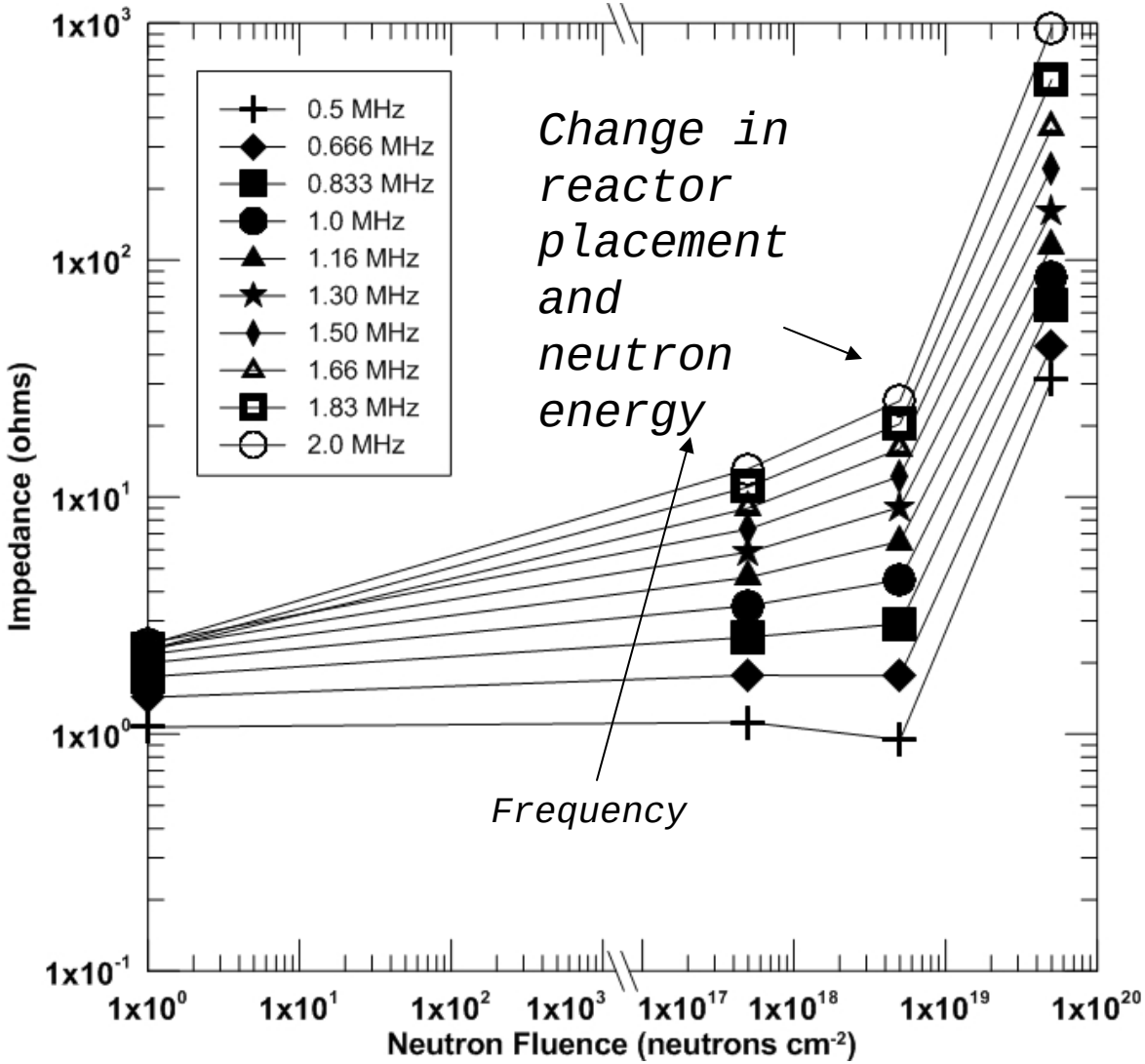
- Sweep through frequencies from 0.5MHz-2.0MHz--even divisions
- Highest frequencies show most pronounced changes





Results and Analysis

Impedance Results

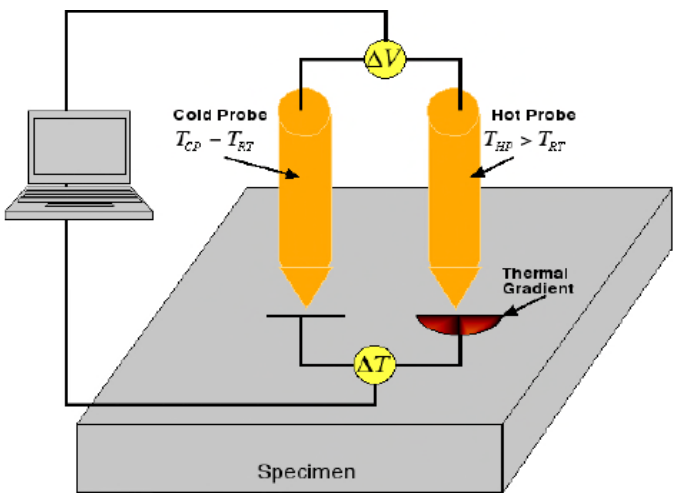


Quantum Mechanical Principle of Thermoelectric Power

Free Electron Model for High Carrier Concentrations

$$S = \frac{\Delta V}{\Delta T}$$

$$S = \left(\pm\frac{k}{e}\right)(27.1)\left(r + \frac{3}{2}\right)\left(\frac{m_e}{h^2}\right)\left(kTn^{\left(-\frac{2}{3}\right)}\right)$$



$$S = S(n, m_e, r)$$

- S Thermoelectric power
- r Scattering parameter
- h Planck constant
- k Boltzmann's constant
- n Free electron concentration
- m_e Effective mass (m^*)

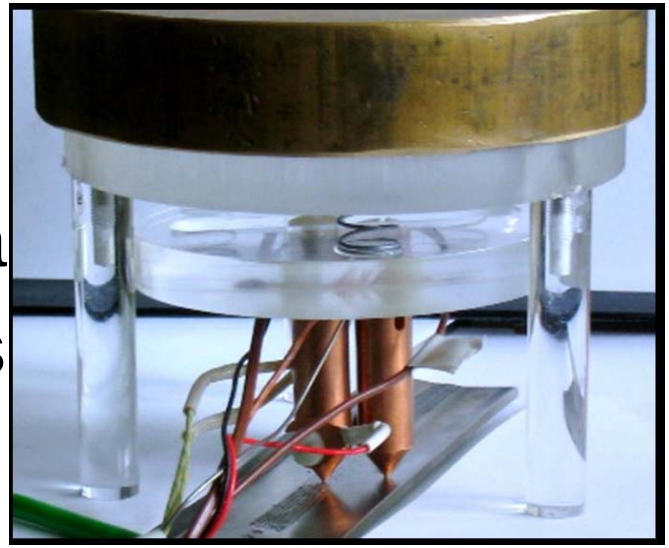
$$m_e = \frac{\hbar^2}{(d^2E/dk^2)}$$

Thermoelectric Power
Seebeck Coefficient

- Determined from ratio of voltage drop to temperature difference across a material

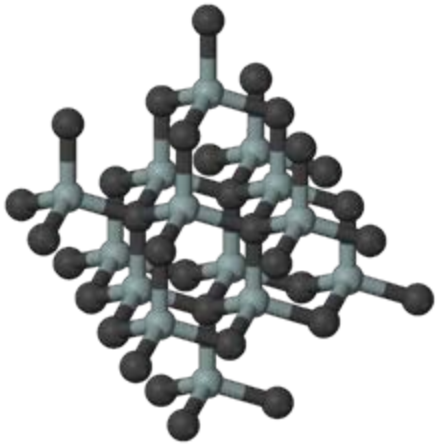
- Difficult to Apply
  - Samples are too sma
  - Conductivity issues

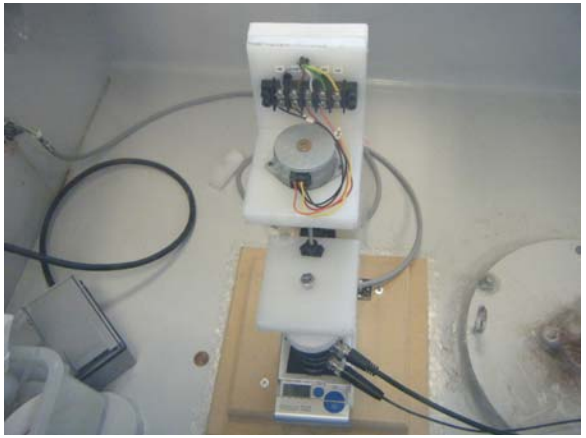
$$S = \frac{\Delta V}{\Delta T}$$

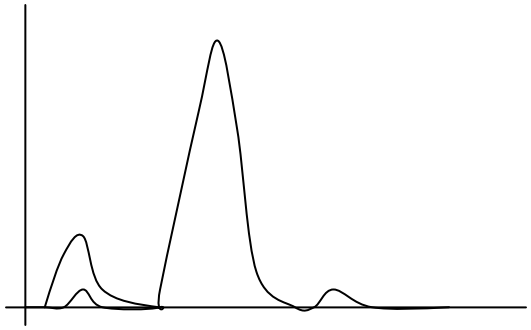


Ultrasonics

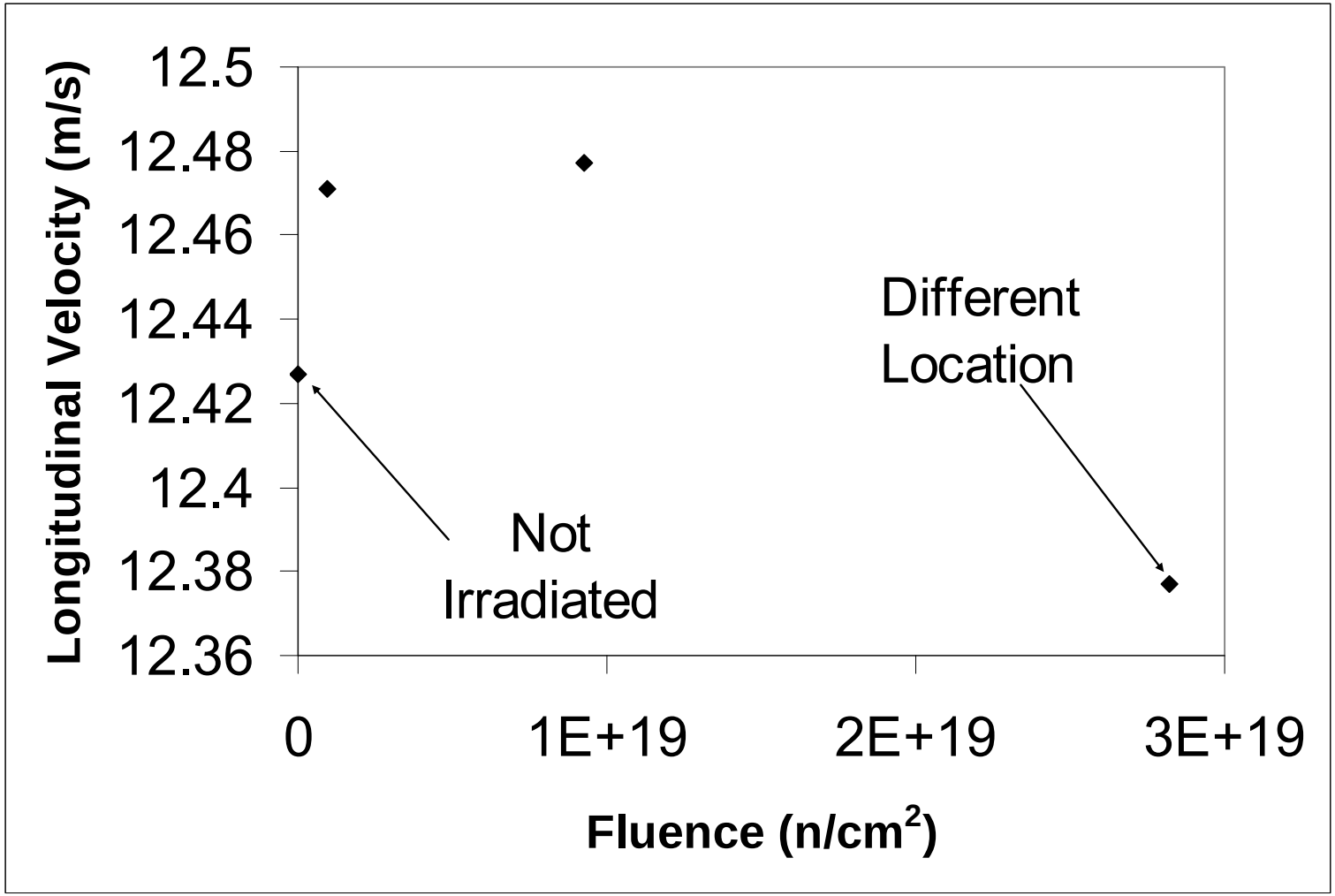
- High frequency waves are used to examine structural changes and bulk defects.
- (f)(λ) = speed of wave propagation, wavelength correlation to microstructure



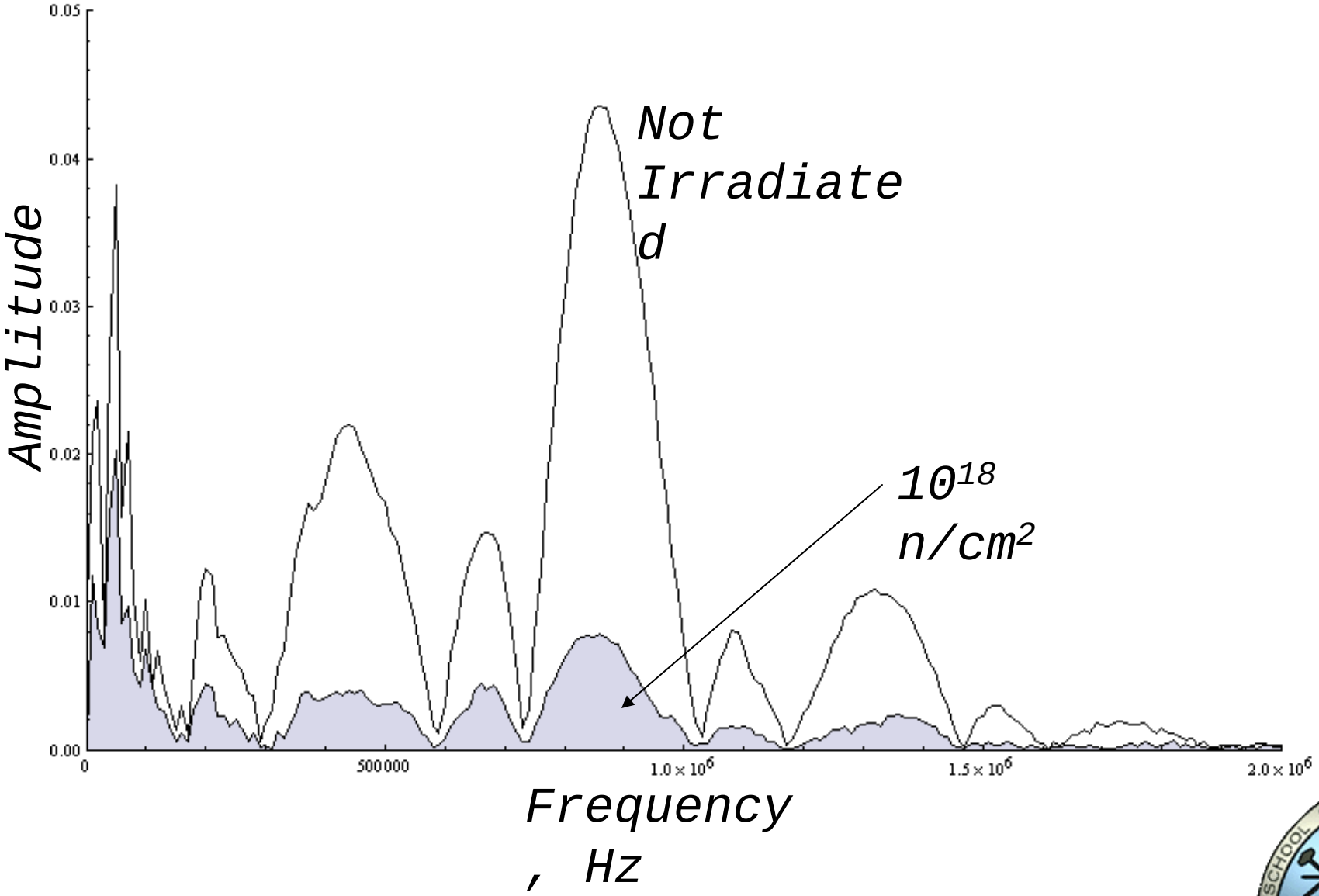




SiC Longitudinal Wavespeed



SiC Resonance Modes



Transmission Electron Microscopy (with X-ray Diffraction Analysis)

- High resolution imaging of structural damage and modification of SiC polytypes.

- Structural defect correlations to QNDE physical property measurements are essential to understand the nature of material damage

Gamma Spectroscopy

Gamma Spectroscopy Measurement Apparatus

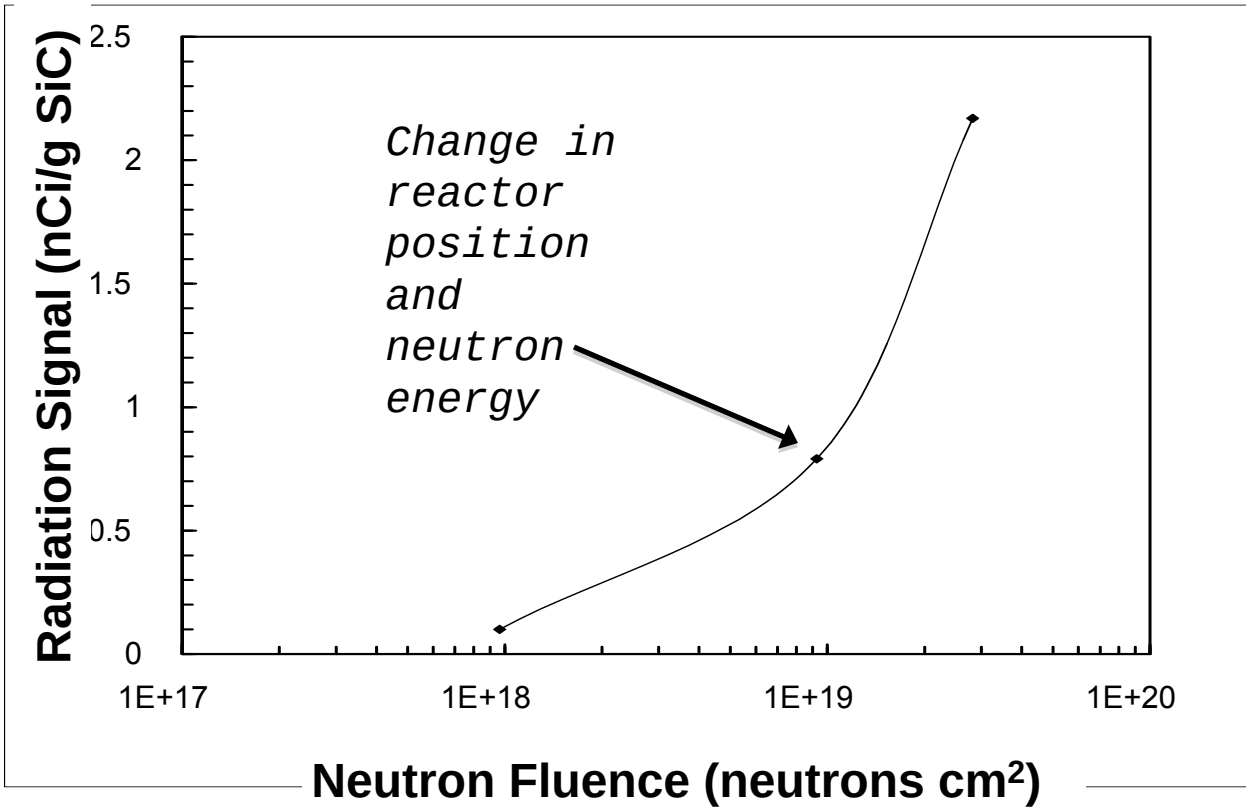
CSM Physics Department

Gamma Spectroscopy Results

- Very low radioactivity
  - Still enough to be measureable
- Primarily Co-60
- Other isotopes observed include: vanadium, chromium, iron, hafnium and antimony
- Increasing neutron fluence leads to increased radioactivity.

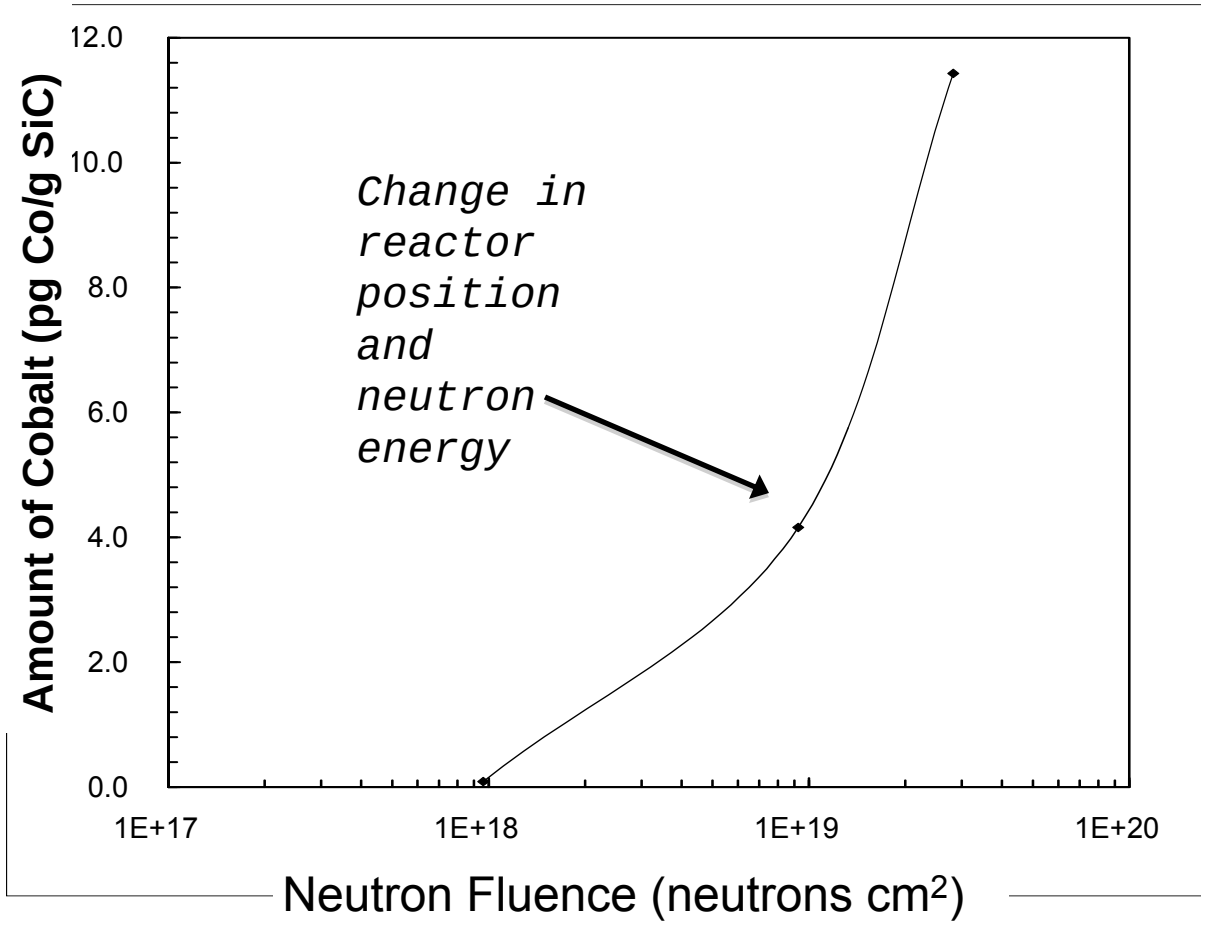
Results and Analysis

Cobalt Radioactivity



Results and Analysis

Increasing Cobalt



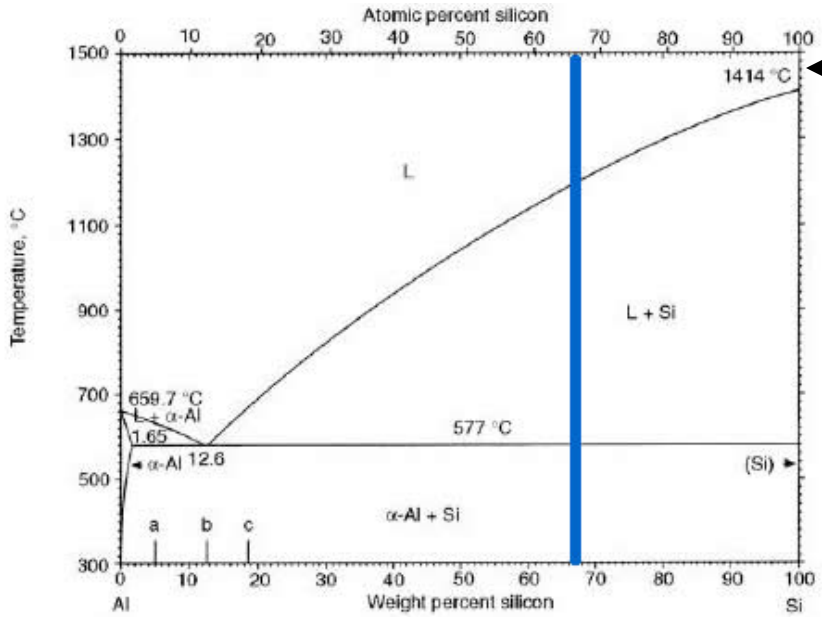
Comparison of Mechanical and Electrical Oscillation

$$F = G\frac{m_1 m_2}{r^2} \qquad F = \frac{qq'}{r^2}$$

Mass on a spring	Circuit containing inductance and capacitance
$\frac{1}{2}mv^2 + \frac{1}{2}kx^2 = \frac{1}{2}kA^2$	$\frac{1}{2}Li^2 + \frac{1}{2}\frac{q^2}{C} = \frac{1}{2}\frac{Q^2}{C}$
$v = \sqrt{\frac{k}{m}}\sqrt{A^2 - x^2}$	$i = \sqrt{\frac{1}{LC}}\sqrt{Q^2 - q^2}$
$v = \frac{dx}{dt}$	$i = \frac{dq}{dt}$
$\int \frac{dx}{\sqrt{A^2 - x^2}} = \sqrt{\frac{k}{m}}\int dt$	$\int \frac{dq}{\sqrt{Q^2 - q^2}} = \sqrt{\frac{1}{LC}}\int dt$
$\sin^{-1}\frac{x}{A} = \sqrt{\frac{k}{m}}\,t$	$\sin^{-1}\frac{q}{Q} = \sqrt{\frac{1}{LC}}\,t$
$x = A\sin\sqrt{\frac{k}{m}}\,t$	$q = Q\sin\sqrt{\frac{1}{LC}}\,t$
$\omega = 2\pi f = \sqrt{\frac{k}{m}}$	$\omega = 2\pi f = \sqrt{\frac{1}{LC}}$
$x = A\sin\omega t$	$q = Q\sin\omega t$

$$Z = \left(R^2 + \left(\omega m - \frac{s}{\omega}\right)^2\right)^{\frac{1}{2}} \qquad Z = \left(R^2 + \left(\omega L - \frac{1}{\omega C}\right)^2\right)^{\frac{1}{2}}$$

Electronic and Elastic Metallography Lab



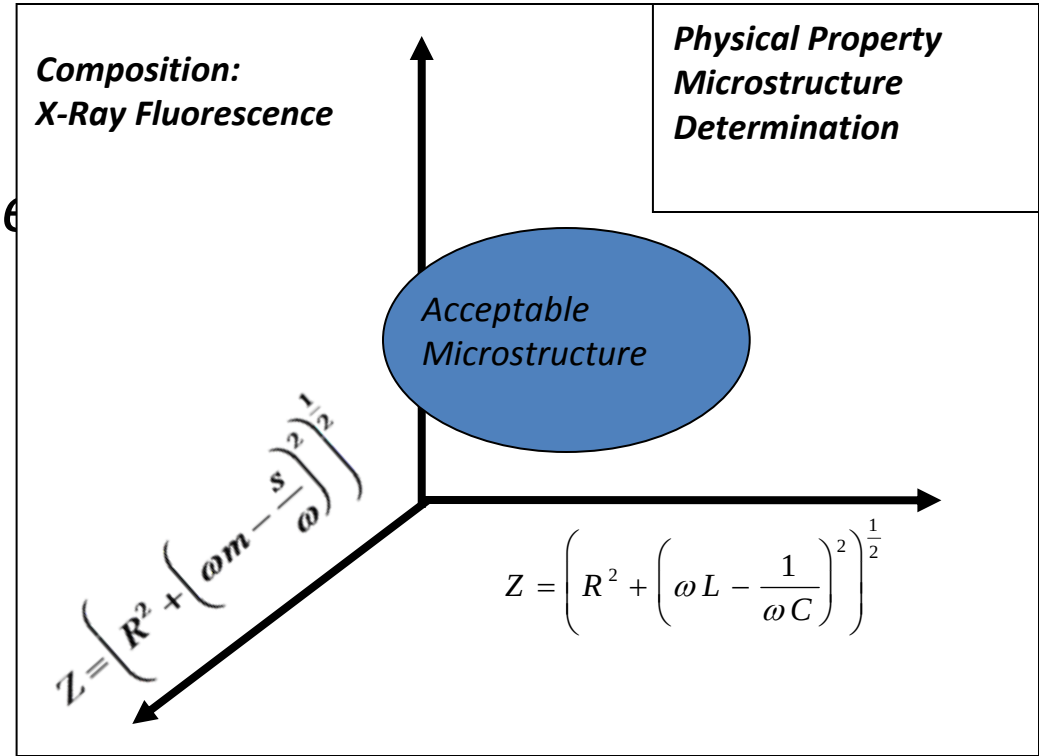
$$P + F = C + 2 + M$$

P = # of phases

F = # of intrinsic variables to describe a microstructure

Intrinsic variables are

J. Jackson 2006



Electronic Impedance

$$Z = \left(R^2 + \left(\omega L - \frac{1}{\omega C}\right)^2\right)^{\frac{1}{2}}$$

Acoustic Impedance

$$Z = \left(R^2 + \left(\omega m - \frac{s}{\omega}\right)^2\right)^{\frac{1}{2}}$$

Conclusions

- Beta SiC could be effectively used as an in-situ reactor core radiation damage monitoring material.

- High frequency impedance results are supported by Hall coefficient and ultrasonic wave analyses and gamma spectroscopy.

- High frequency impedance analysis (with harmonic and frequency analyses) supported by elastic wave analyses is very promising QNDE technology for in-situ

Future Investigations

- TEM and XRD to visually determine changes in material structure relative to QNDE measurements.

- SiC with cadmium coating studies to separate the effects of fast and thermal neutrons (nuclear transmutation and lattice damage).

- QNDE assessment for irradiated

Potential Problems

- Further investigation needs to be conducted regarding the effect of fast neutrons on sensor reliability. A cadmium coating on the SiC will allow the magnitude of this issue to be studied.

- Monitored properties may not be linear with irradiation damage at high fluence levels.

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