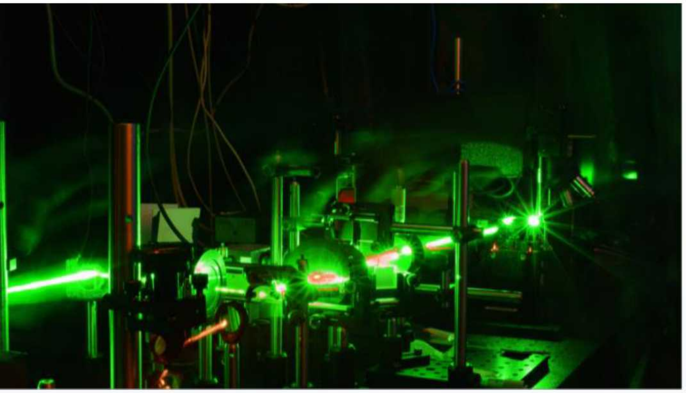
SAND2020-2772C

Tail… and Acoustic Properties through Ion Beam Modification

Khalid Hattar

Ion Beam Lab and Center for Integrated Nanotechnologies

at Sandia National Laboratories



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Combining ion irradiation and pump-probe laser testing provides a unique and rapid means of producing new avenues for thermal and mechanical studies

Collaborators:

- Sandia: D. Buller, T. Beechem III, C. Saltonstall, E. Ziade, R.C. Choens, C.A. Taylor, N.M. Heckman, M.D. Ingraham, D. Robinson, B.L. Boyce
- External: Ethan Scott, Cody Dennett, Michael Short, Patrick Hopkins, Mark Goorsky, Keivan Esfarjani, Jeffrey Braun, Christina Rost, John Gaskins, Mehrdad Fazli, Claire Ganski, Chao Li, Tingyu Bai, Yekan Wang

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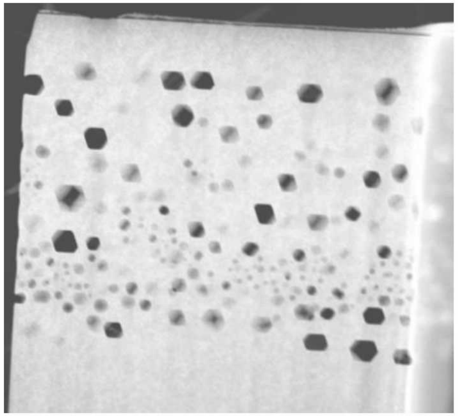
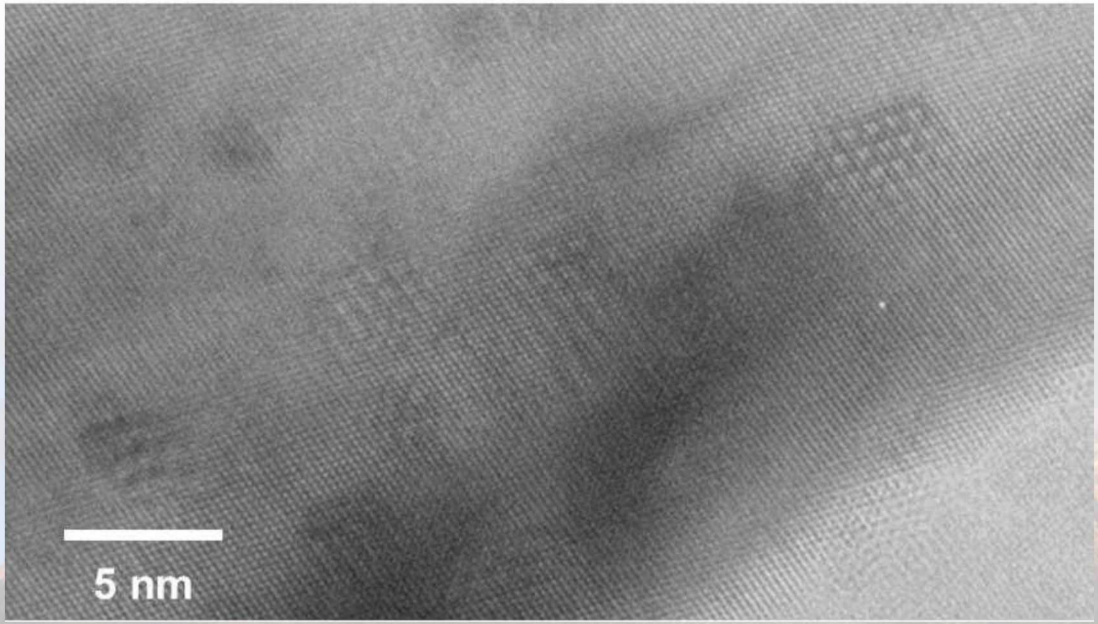
Why Focus on Radiation Interactions?

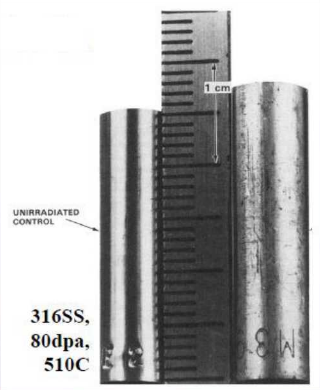
Ion implantation offers a unique way to tailor material properties

- **Electrical Properties**
  - Local doping of semiconductors revolutionized our world
- **Structural Properties**
  - Void formation and swelling
  - Defect clustering and embrittlement
- **Thermal Properties**
  - Thermal conductivity reductions
  - Reductions and *increases* in thermal boundary conductance



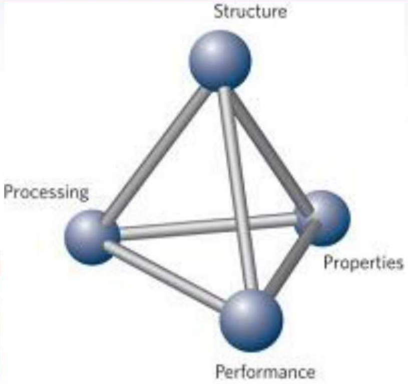


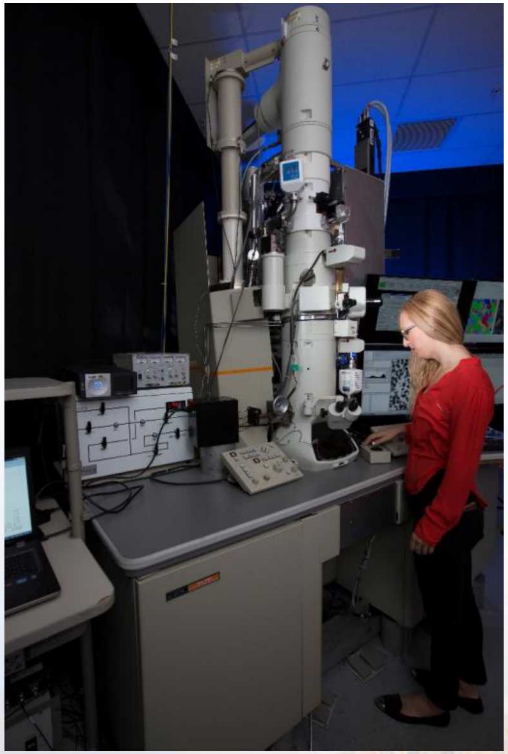
Radiation damage can vastly improve or devastate our lives depending on how it is wielded.



Major Limitation of In-situ Electron Beam Characterization of Radiation Damage

I. **Excellent capability to corelate the processing – structure effects resulting from ion irradiation or implantation**
   I. **Either simulate real world radiation environments**
   II. **Create far-from-equilibrium tailored microstretches**





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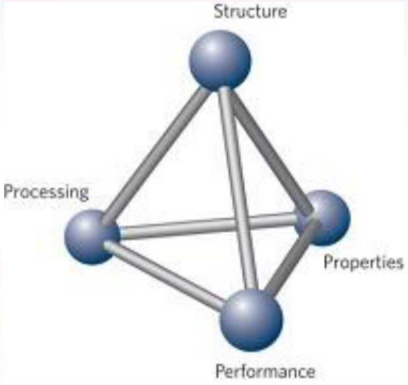

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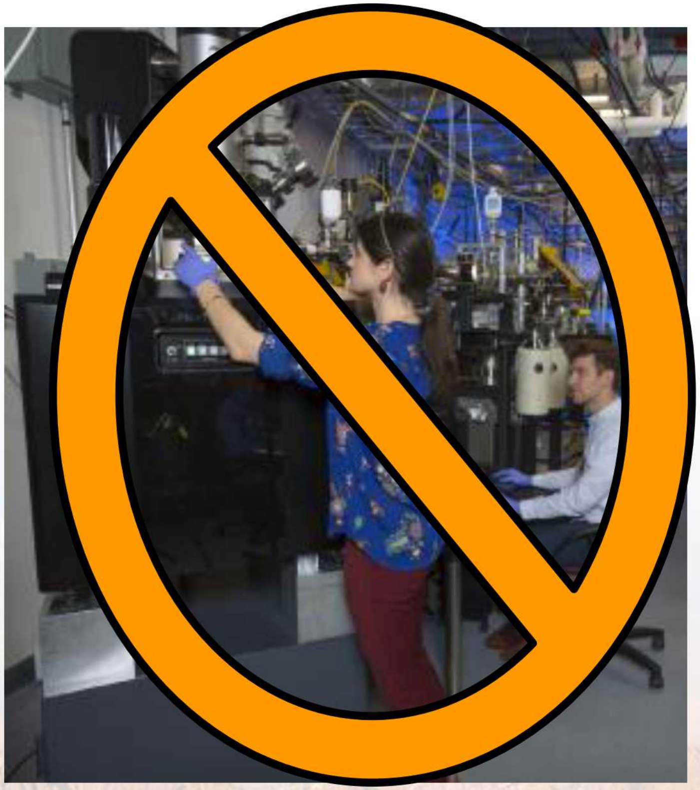
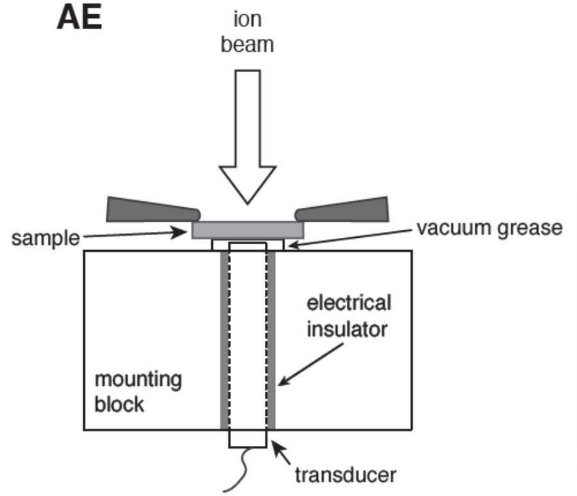


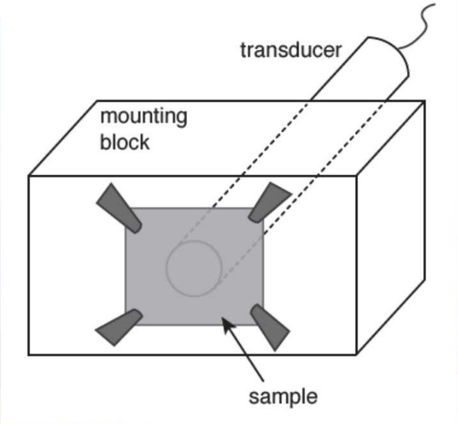
Provides minimal, if any, property information

Can We Develop a Simple Non-Intrusive Way to Listen to Sample During Ion Bombardment?

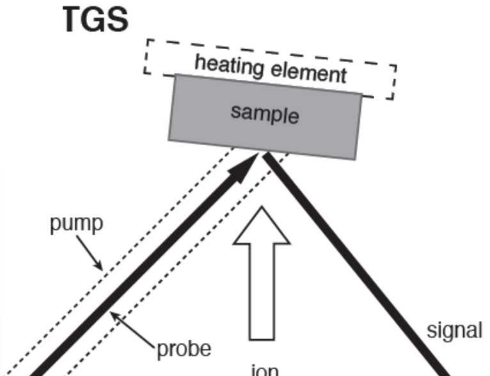
Collaborator: C.A. Dennett, R.C. Choens, C.A. Taylor, N.M. Heckman, M.D. Ingraham, D. Robinson, B.L. Boyce, M.P. Short

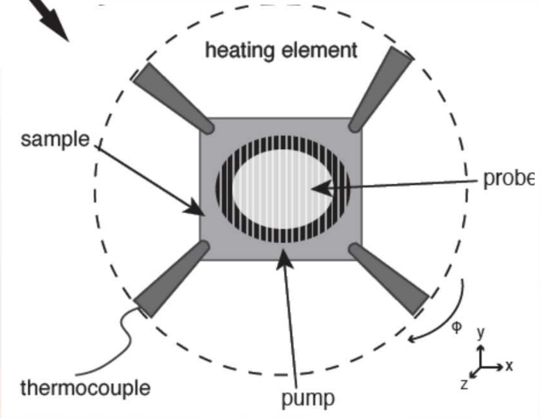
Passive Listening: Acoustic Emission





Active Listening:
Pump-Probe Laser Techniques





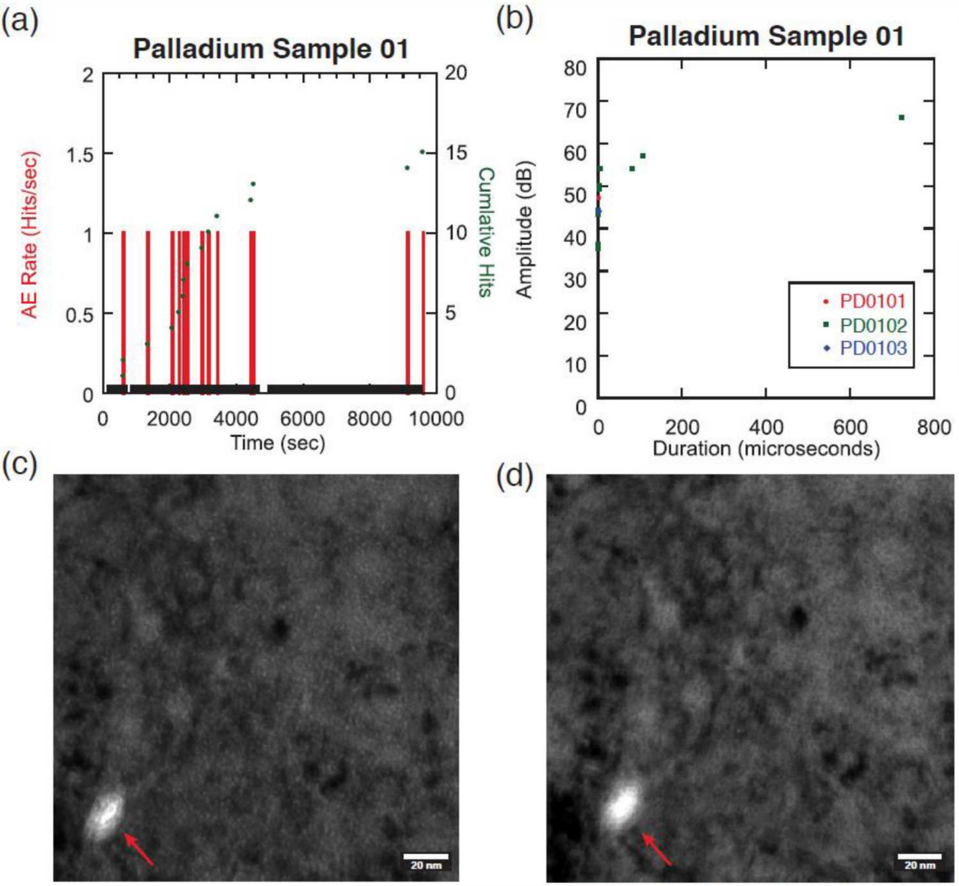
It is better to listen than assume!
It is better to listen before acting!

Dennett, Cody A., et al. "Listening to Radiation Damage In Situ: Passive and Active Acoustic Techniques." *JOM* 72.1 (2020): 197-209.

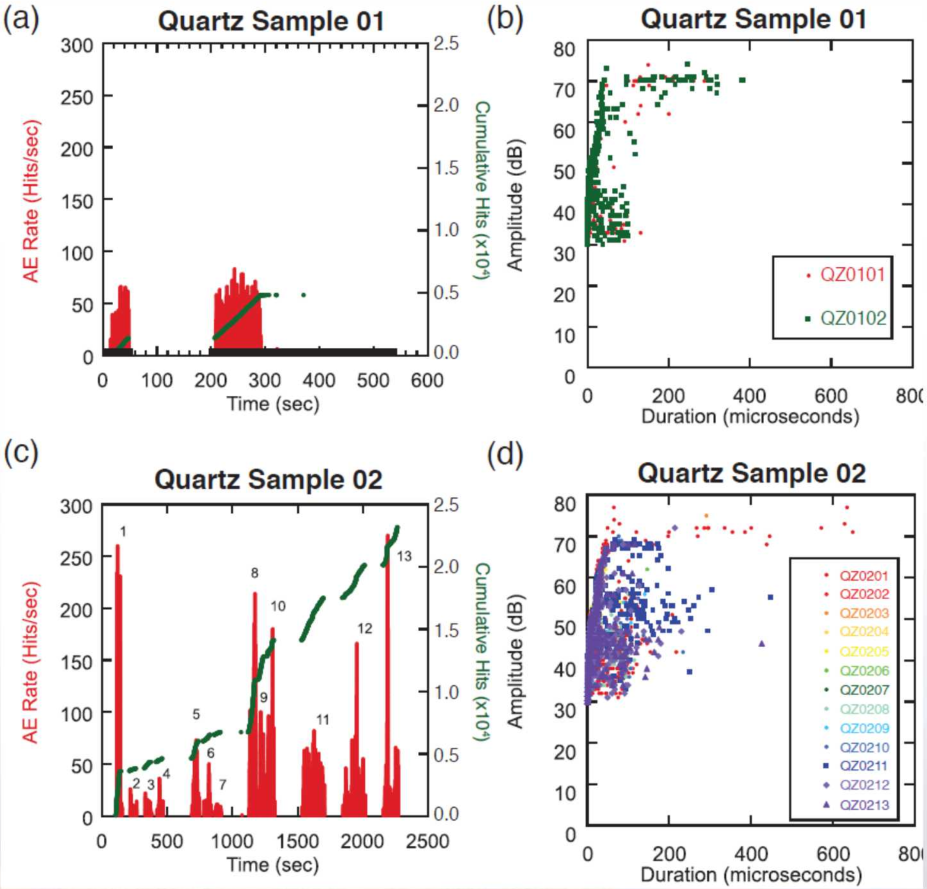
Acoustic Emission Response to 2 MeV He^+

Collaborator: C.A. Dennett, R.C. Choens, C.A. Taylor, N.M. Heckman, M.D. Ingraham, D. Robinson, B.L. Boyce, M.P. Short

Metals



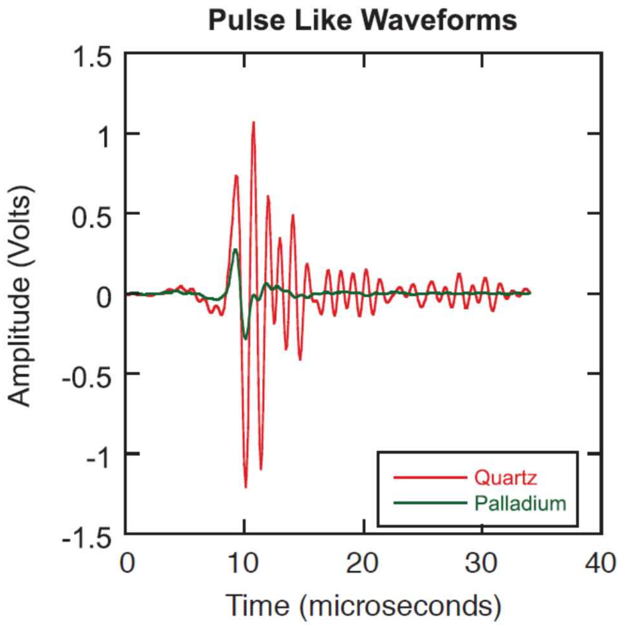
Ceramics

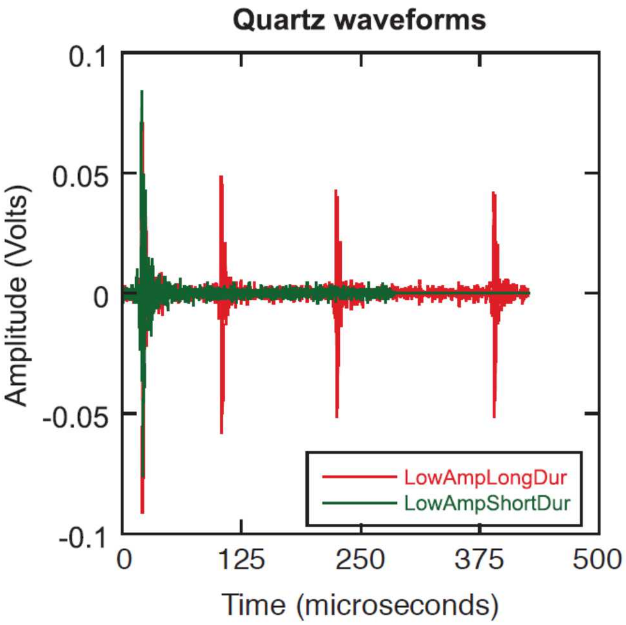


AE can be applied to any material!

Tells when catastrophic change occurs during ion beam modification

Individual Events





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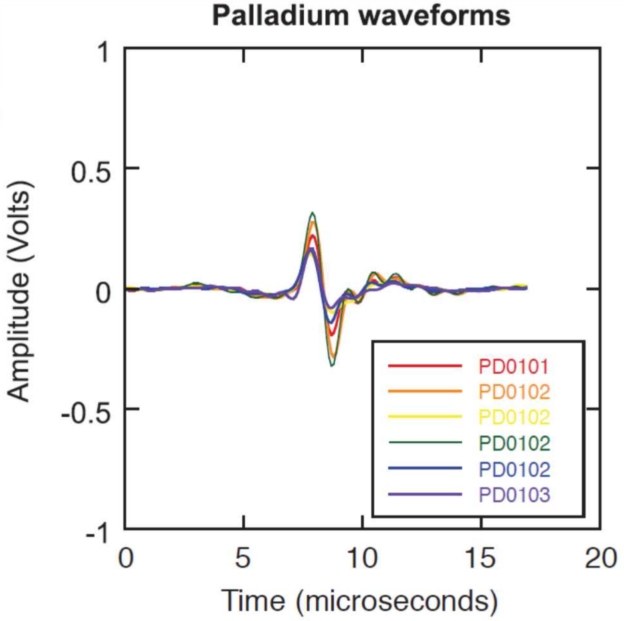
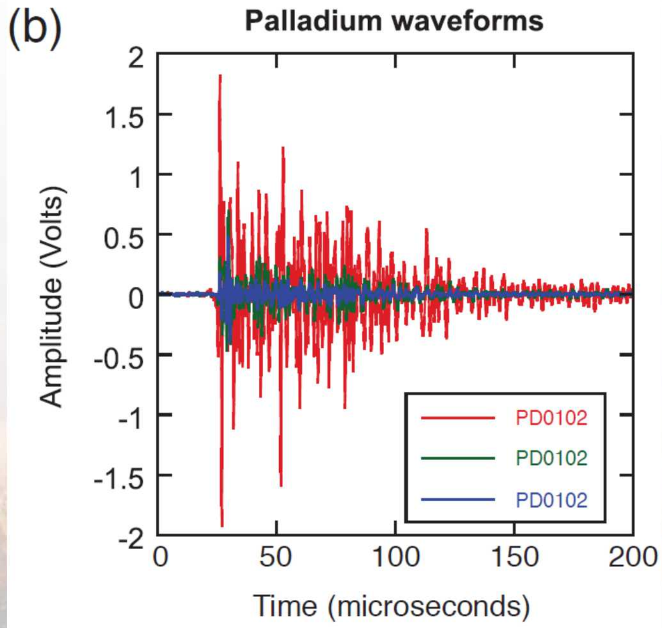
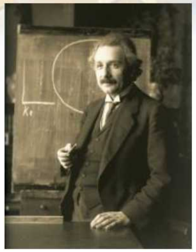
Difference sound resulting from each acoustic event that can be associated with materials catastrophic response.

Dennett, Cody A., et al. "Listening to Radiation Damage In Situ: Passive and Active Acoustic Techniques." ***JOM*** **72.1 (2020): 197-209.**





Lasers and Particle Accelerators through the Years



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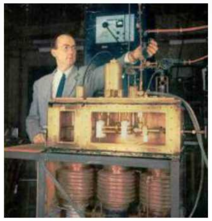
Einstein proposes idea of stimulated emission

Fabrikants applies theory to amplify radiation

1939

1951

Kastler proposes method of optical pumping



C. Townes develops microwave “MASER”

1953

1960

T. Maiman (Hughes Research Lab,) creates first operational laser

Invention of semi-conducting diode laser

1962

1982

First CD player

Fiber-optic laser used in brain cancer surgery

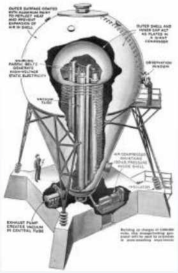
2008

2010

Movement towards fusion (192 laser beams compress deuterium/tritium)

1895

Rontgen discovers x-rays with a cathode ray tube



Van de Graaff generator developed

1929

1929-1930

Cyclotron developed by E. Lawrence

Cockroft and Walton accelerate protons to 800 keV with the CW generator

1932

1945

E. McMillan constructs first electron synchrotron

M. Oliphant designs and builds the first proton synchrotron

1952

1962

Establishment of Stanford Linear Accelerator

These technologies have been evolving for over a century, opening many new fields of research and life

*Images from Wikipedia

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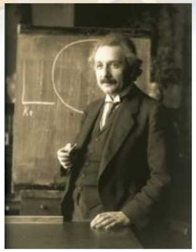
Lasers and Particle Accelerators through the Years



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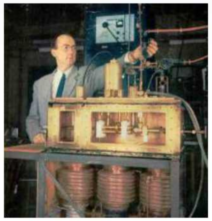

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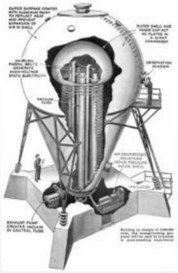

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What if they were converging historical paths?

*Images from Wikipedia

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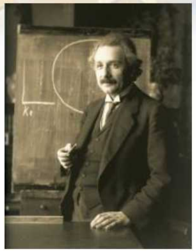

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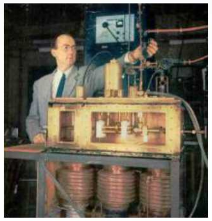

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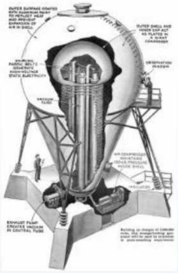

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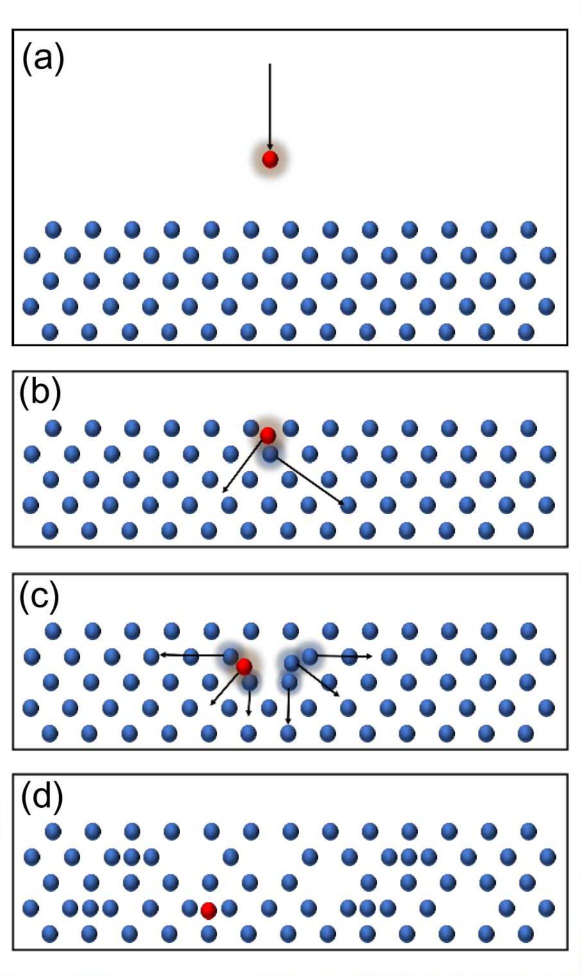
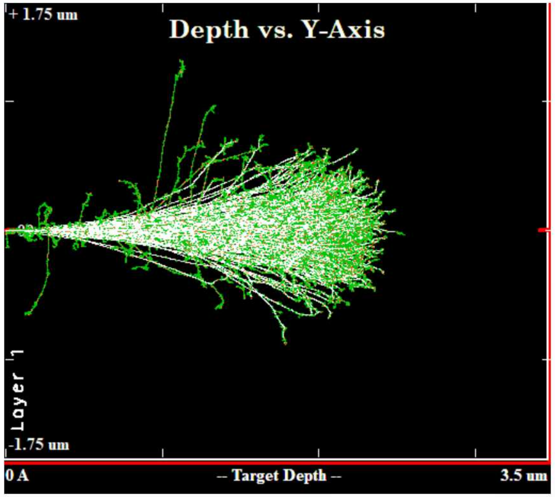
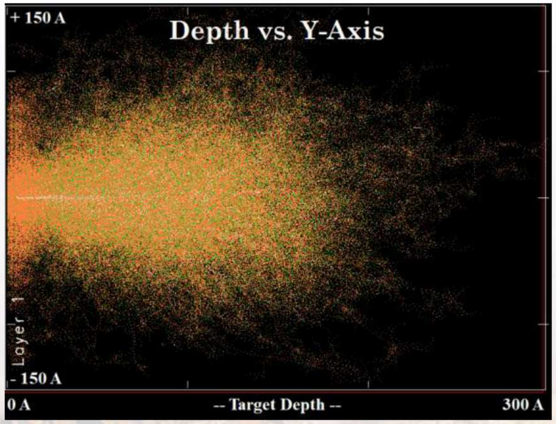
Our efforts to align Ion Beam Modification and Laser –based Pump Probe Analysis

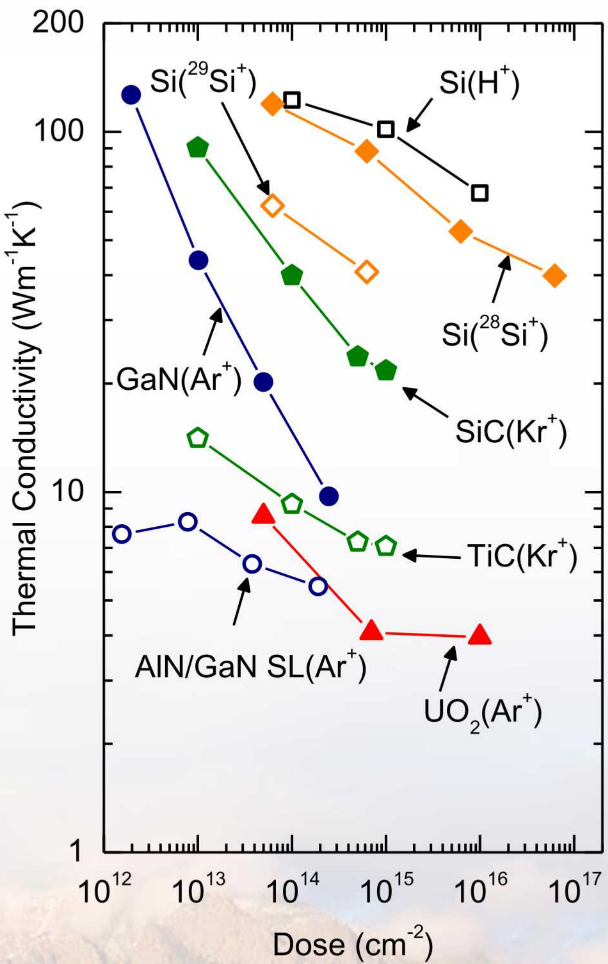
*Images from Wikipedia

How do Ion Beams Interact with a Lattice?









Scott et al, *PRM*, **2**, 095001 (1977)

Energetic particles create recoil cascades, yielding vacancies, interstitials, Frenkel pairs as well as extended defects

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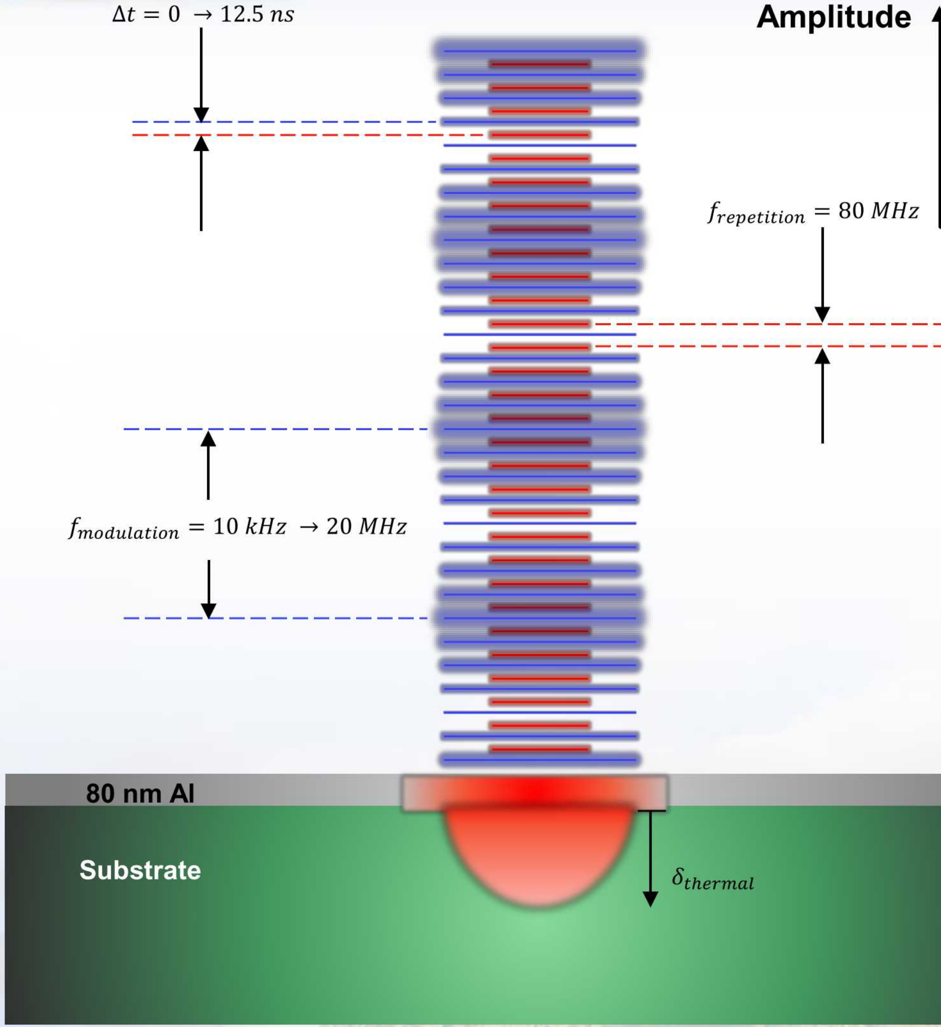
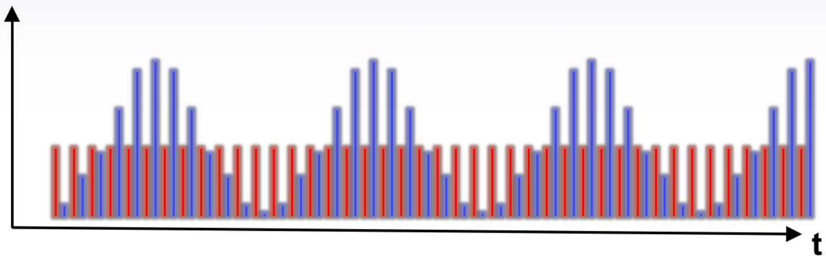
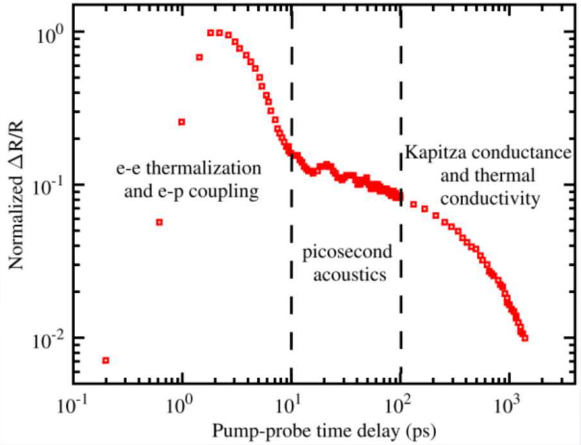
How Can We Quantify the Thermal Effects?





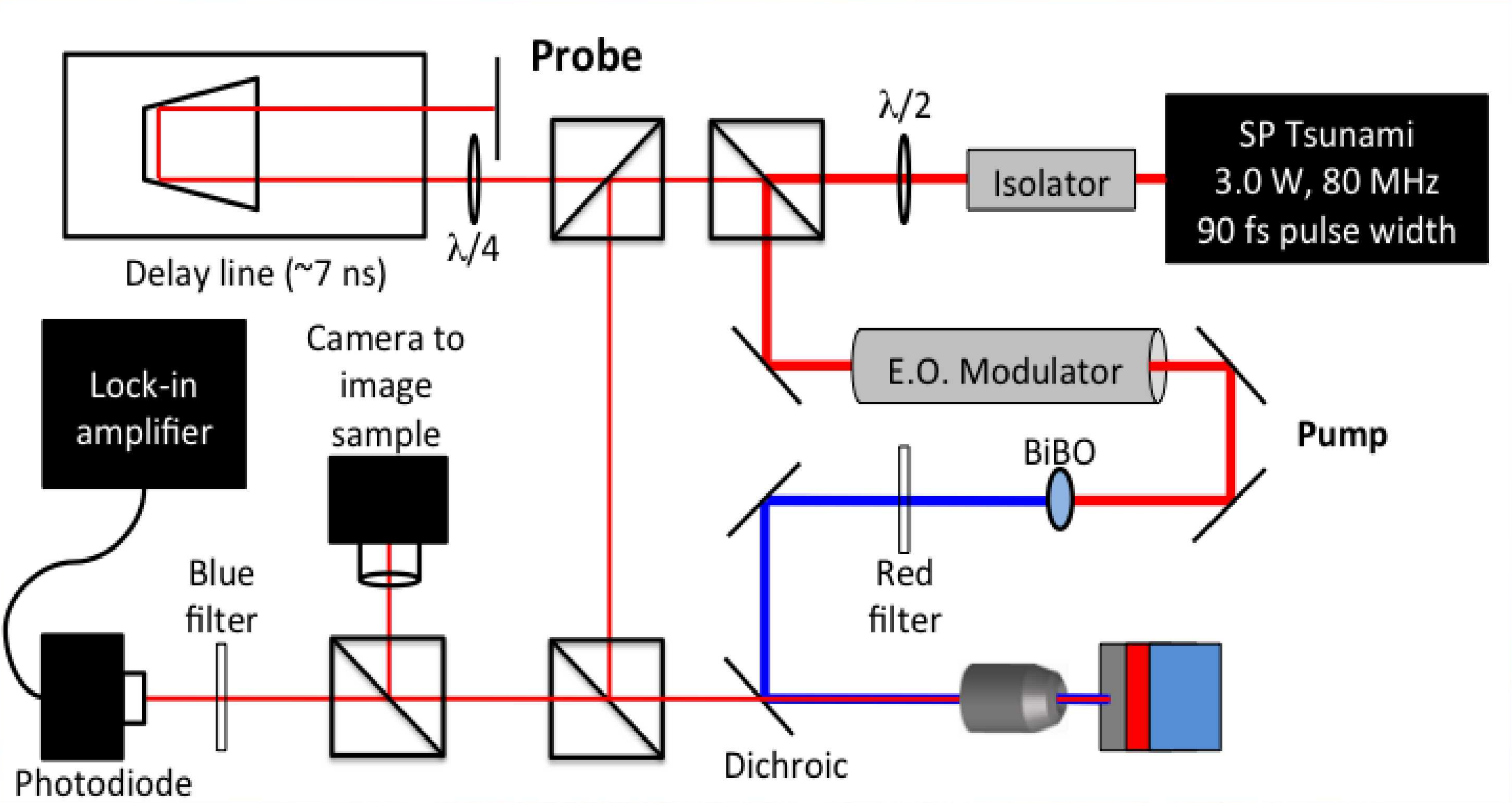


Giri *et al.*, *JAP*, 117, 105105 (2015)

TDTR is a laser pump probe technique with excellent temporal and spatial resolution

A Typical Time-domain Thermoreflectance (TDTR) Setup

Collaborator: Ethan A. Scott, Patrick Hopkins, Mark Goorsky, Chao Li, Tingyu Bai, Claire Ganski



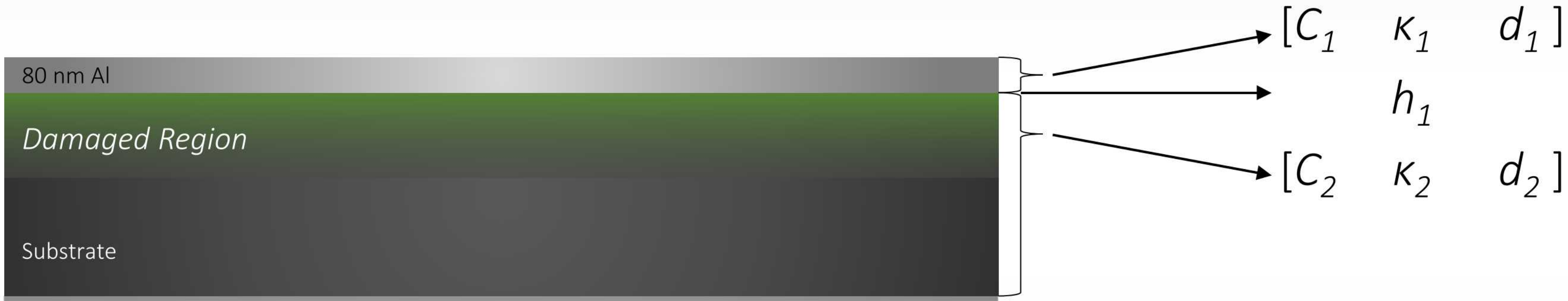
Hopkins *et al.*, *ISRN Mechanical Engineering, 682586 (*2013)

Properties of TDTR

I. Sub-picosecond temporal resolution
II. Spot sizes as low as ~1 μm
III. Modulation frequencies: 10 kHz – 20 MHz

What does TDTR Measure?

Collaborator: Ethan A. Scott, Patrick Hopkins, Mark Goorsky, Chao Li, Tingyu Bai, Claire Ganski

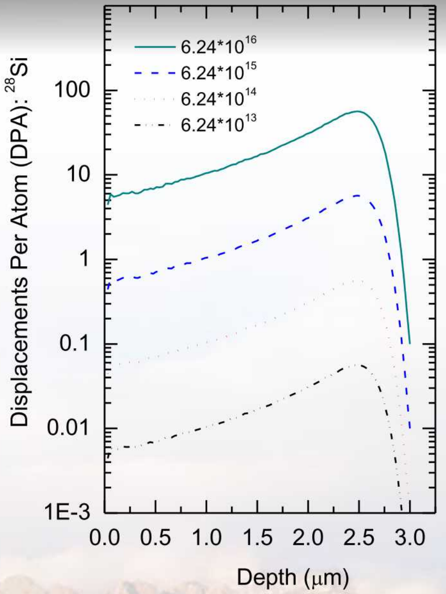


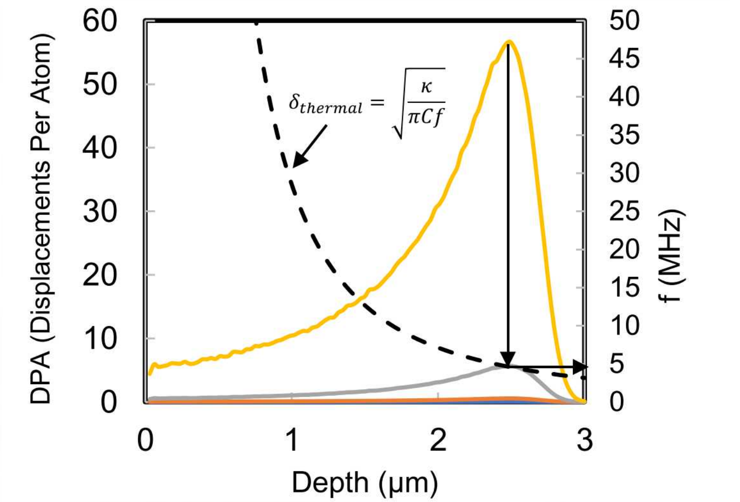
J. F. Ziegler
U.S.N.A.
Annapolis, MD, USA

J. P. Biersack
Hahn-Meitner Inst.
Berlin, Germany

Significant contributions by Helmut Paul (Linz), Roger Webb (Surrey), Xiao Yu (Beijing)

(c) 1984,1989,1998, 2008, 2012 by J. F. Ziegler, M.D. Ziegler, J. P. Biersack (SRIM.com)





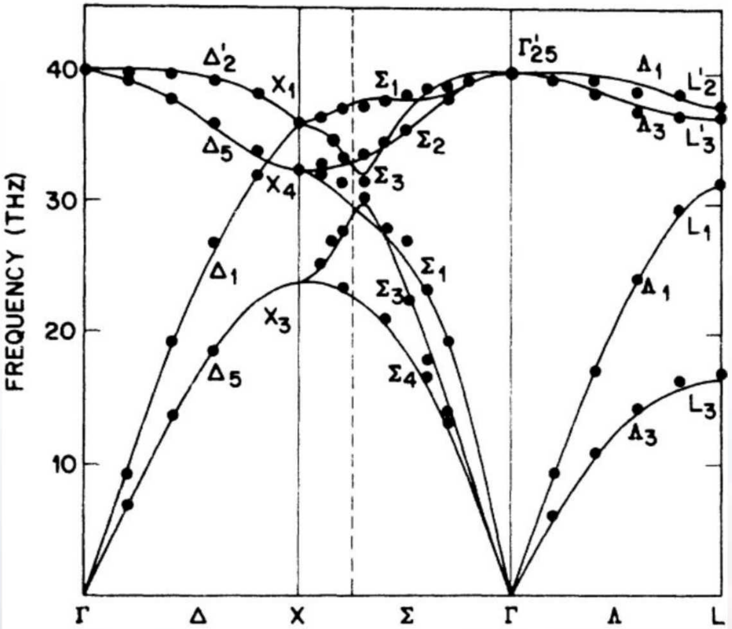
The frequency of modulation controls thermal penetration depth that can be tailored to match the ion implantation depths

Avenue for Exploration: Straining a Lattice through Implantation

Collaborator: Ethan A. Scott, Patrick Hopkins, Mark Goorsky, Chao Li, Tingyu Bai, Claire Ganski

Can we use ion implantation to isolate scattering paramters?

I. **Klemens' model for defect scattering**
    I. Scattering cross section combines effects of mass impurities and strain



Weber, *PRB*, **15**, 4789 (1977)

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

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

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

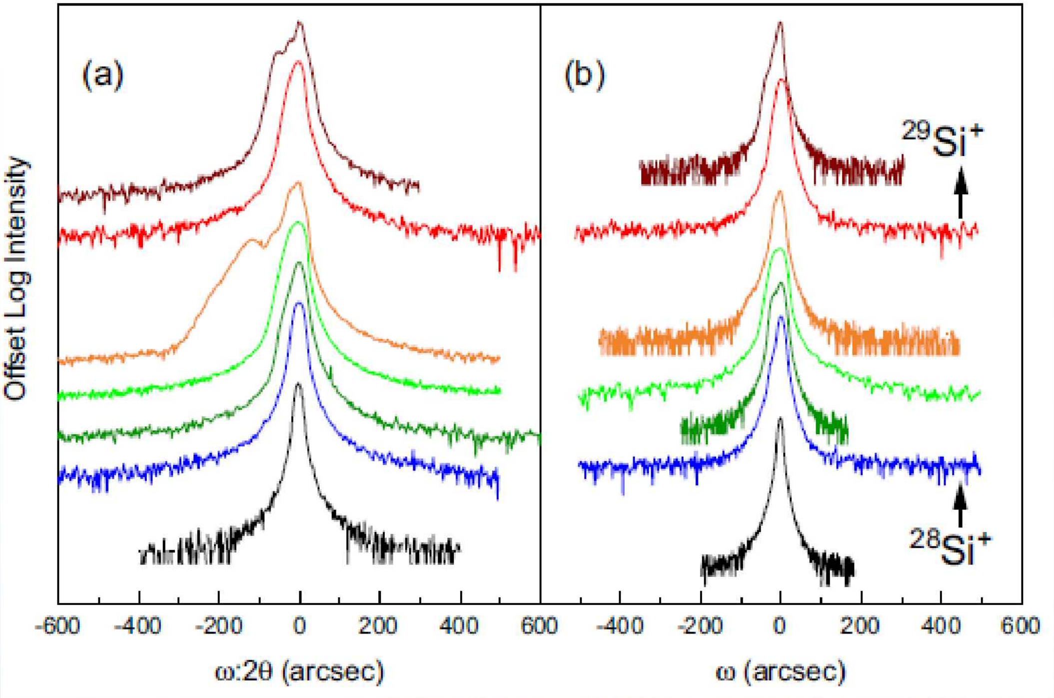
Ion irradiation in combination with TDTR can be used to experimentally investigate classical theories

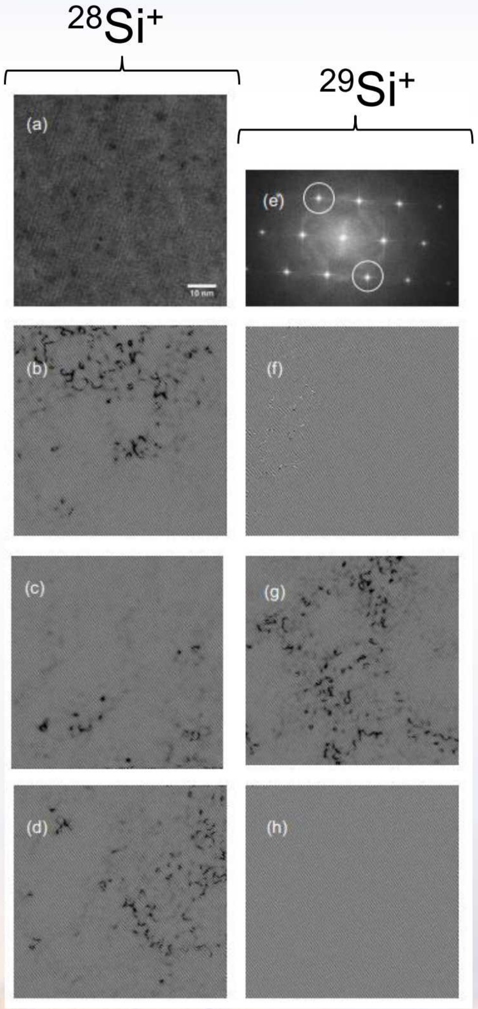
Implanting Isotopes to Create Strain

Collaborator: Ethan A. Scott, Patrick Hopkins, Mark Goorsky, Chao Li, Tingyu Bai, Claire Ganski

$^{28}Si^{+}$, $^{29}Si^{+}$

I. **Ions implanted at 3.75 MeV, up to 6.24(10^{16}) cm^{-2}**
   I. Negligible mass difference
   II. Large measured difference in strain





Implanting isotopes induces additional localized strain with negligible mass differences between ions

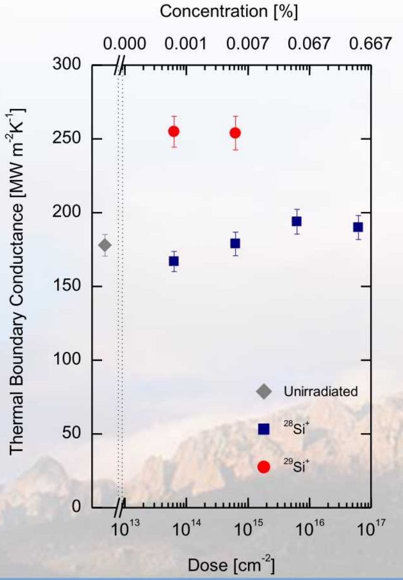
Scott et. al, *PRM,* 2, 095001 (1977)

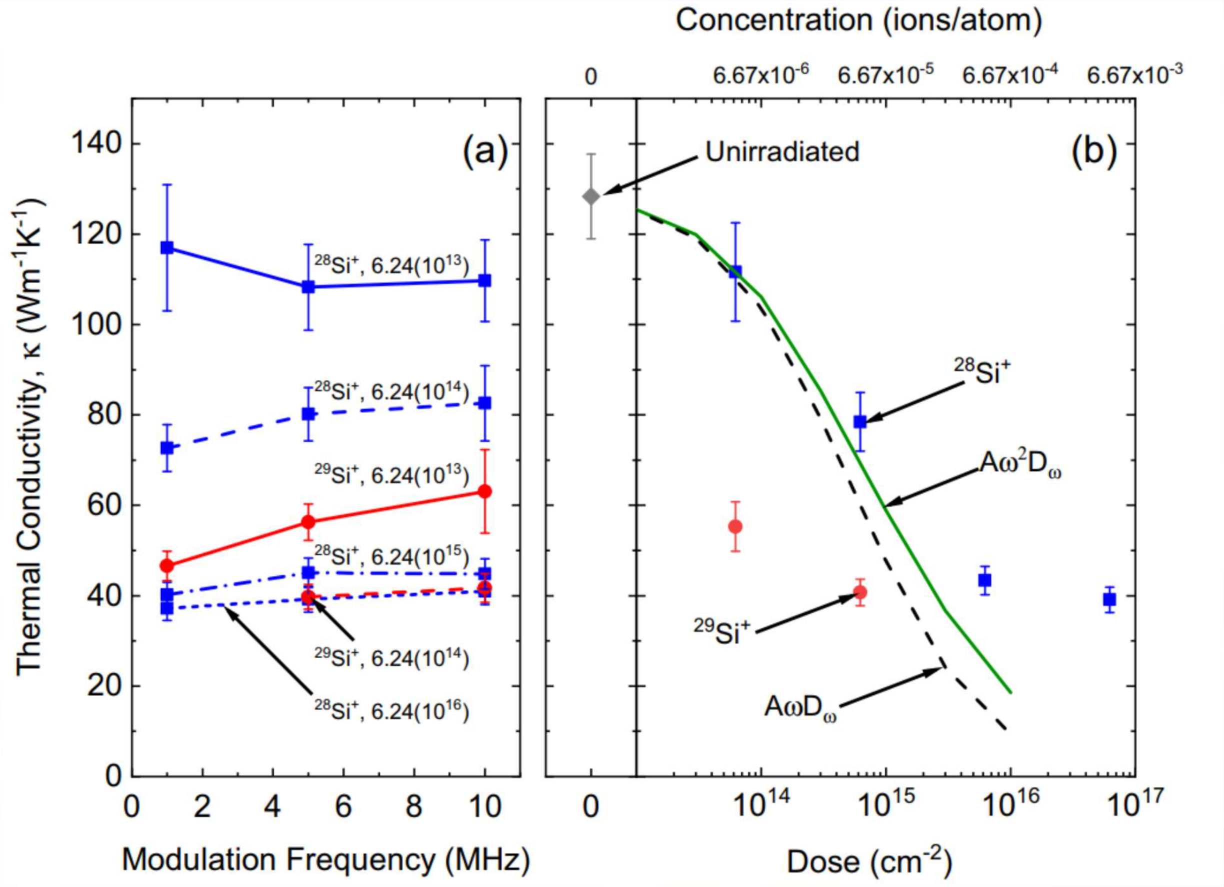
Isotopic Sensitivity

Collaborator: Ethan A. Scott, Patrick Hopkins, Mark Goorsky, Chao Li, Tingyu Bai, Claire Ganski

TDTR Findings

I. **Thermal Conductivity**
    I. Negligible frequency dependence
    II. Reduction as a function of dose
    III. Reduction with larger isotope
II. **Thermal Boundary Conductance**
    I. Increase with dose
    II. Increase with isotope size





Reductions in thermal conductivity cannot be explained by ion mass, rather, increased lattice strain

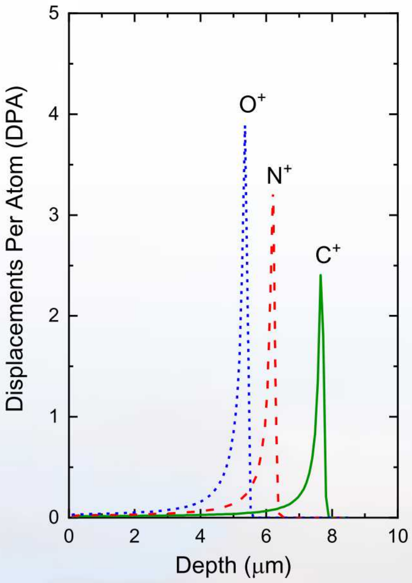
Diamond Doping

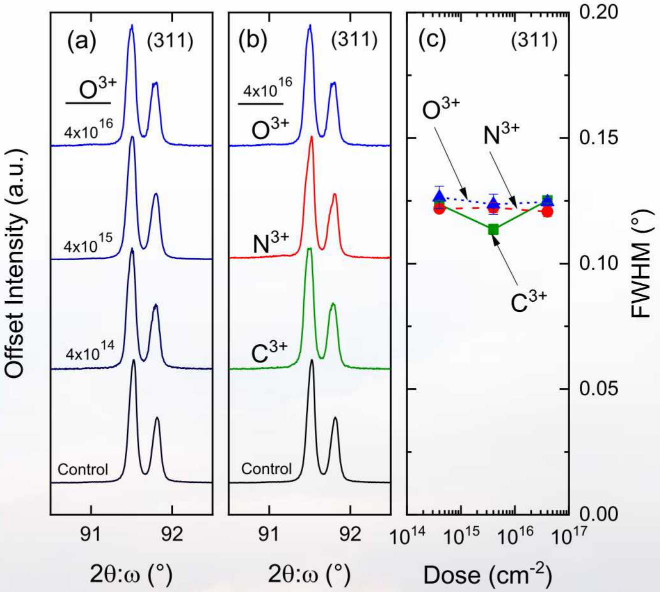
14	15	16
6 **C** carbon 12.011 [12.009, 12.012]	7 **N** nitrogen 14.007 [14.006, 14.008]	8 **O** oxygen 15.999 [15.999, 16.000]

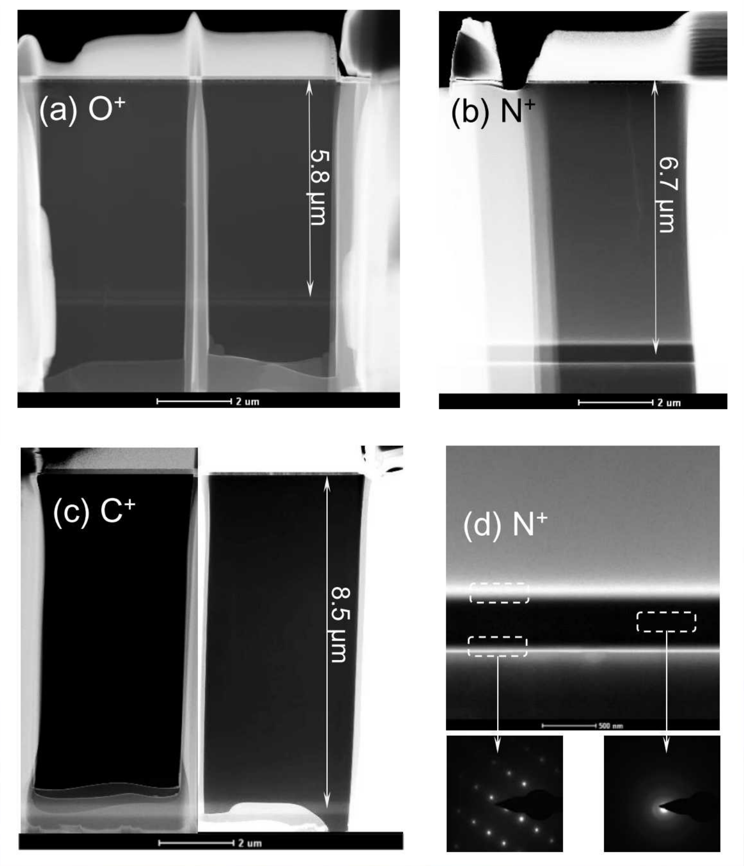
Collaborator: Ethan A. Scott, Patrick Hopkins, Mark Goorsky, Chao Li, Tingyu Bai, Claire Ganski

Diamond Implanted with C^{3+}, O^{3+}, N^{3+}

I. **16.5 MeV, $4(10^{13}) – 4(10^{16})$ cm^{-2}**
   I. Longitudinal straggle more spatially localized
   II. Projected range produces amorphous bands
   III. Lesser strain produced compared to Si implant







Ion irradiating polycrystalline diamond allows for heavier masses, and lesser induced strain

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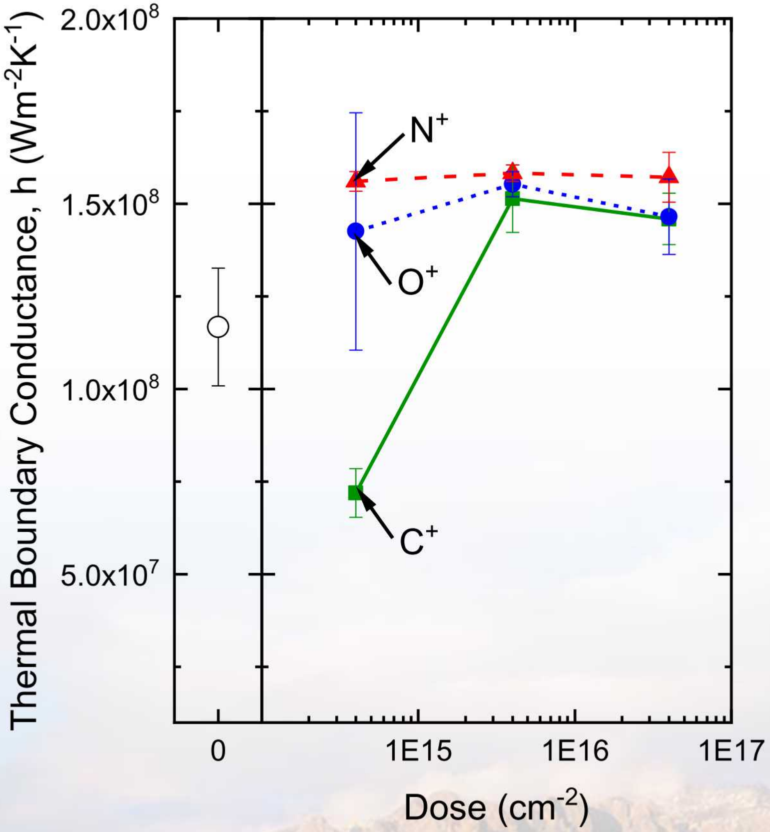
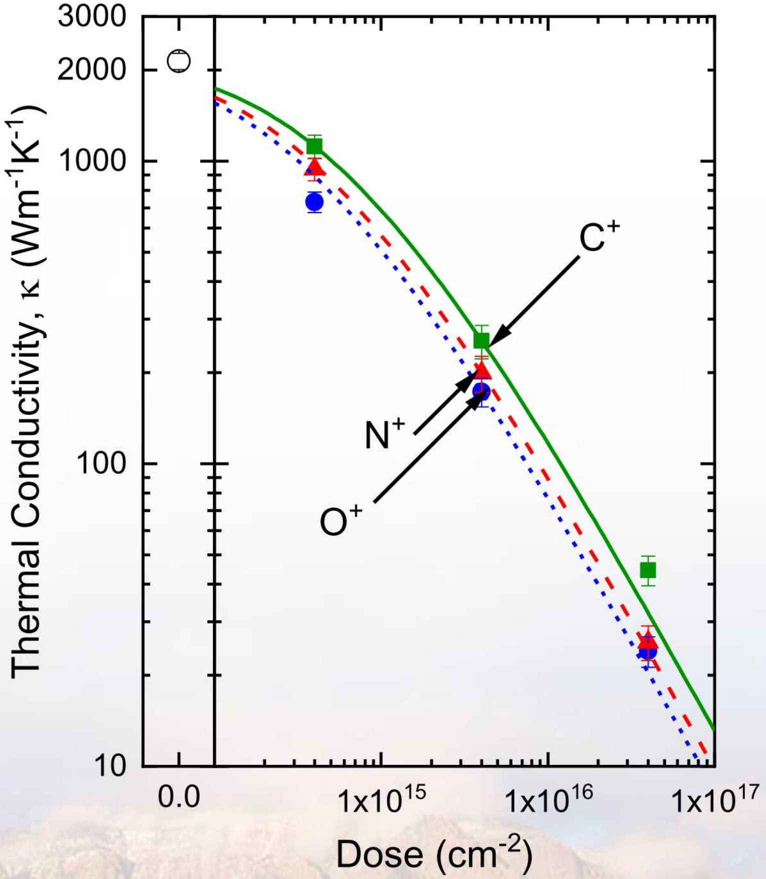
Thermal Degradation of Diamond

Collaborator: Ethan A. Scott, Patrick Hopkins, Mark Goorsky, Chao Li, Tingyu Bai, Claire Ganski

Orders of Magnitude Reduction in $\kappa_{diamond}$

I. **2000 → 20 W m^{-1} K^{-1}**





TBC again *increases*, whereas thermal conductivity decreases by orders of magnitude

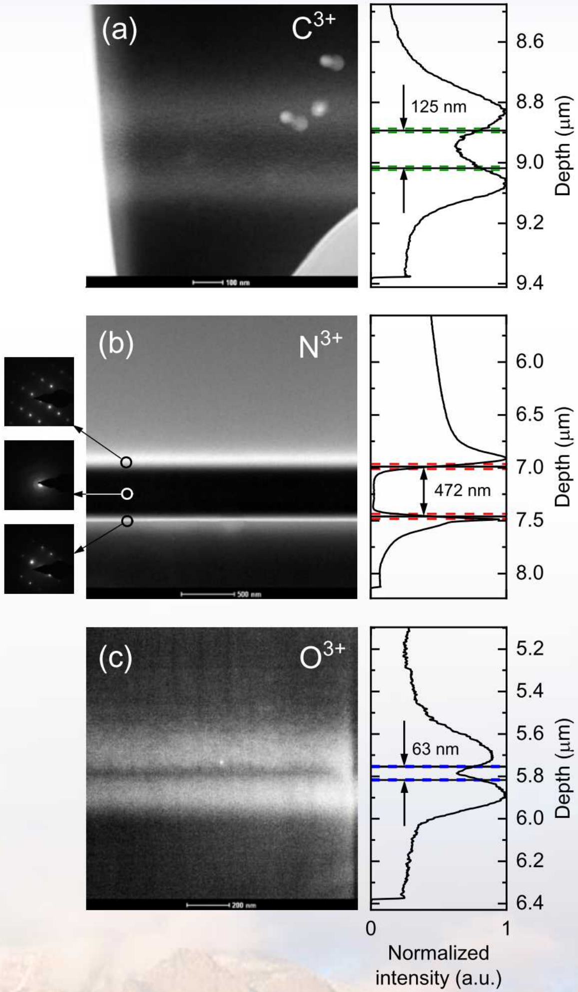
Sandia National Laboratories

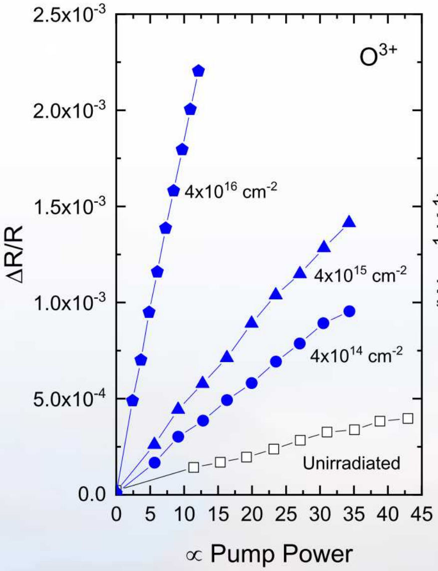
Investigating the Amorphous Band

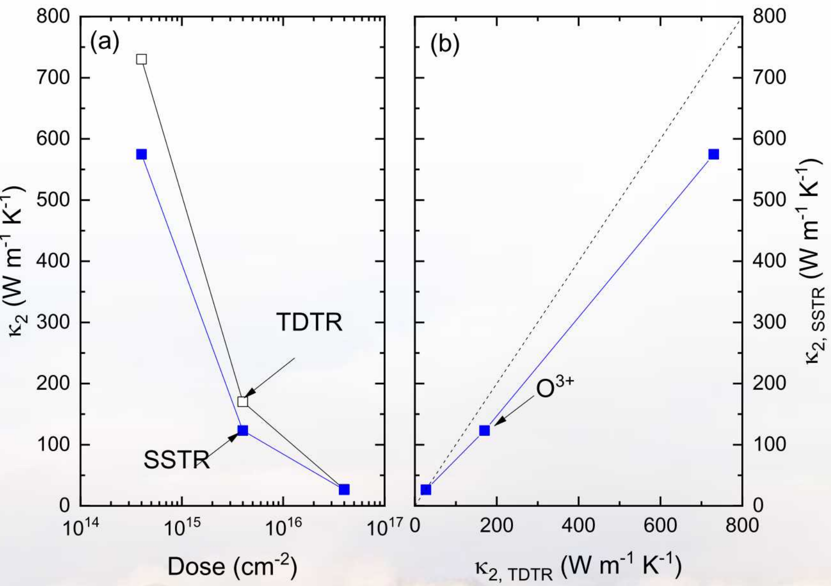
Collaborator: Jeff Braun, Mark Goorsky, Chao Li, Tingyu Bai, Claire Ganski

Steady State Thermoreflectance (SSTR)

I. **Advancements from TDTR**
   I. Thermal penetration depth is limited in TDTR
   II. SSTR can probe further, allowing for analysis of amorphous region
      I. In a 2 layer model, SSTR measures a lower thermal conductivity







Development of a new technique, SSTR, is allowing for thermal analysis deeper into irradiated materials

Creating and Characterizing *Unique* Defect Structures

Implanting Noble Gases

I. **Crystalized "bubble"**

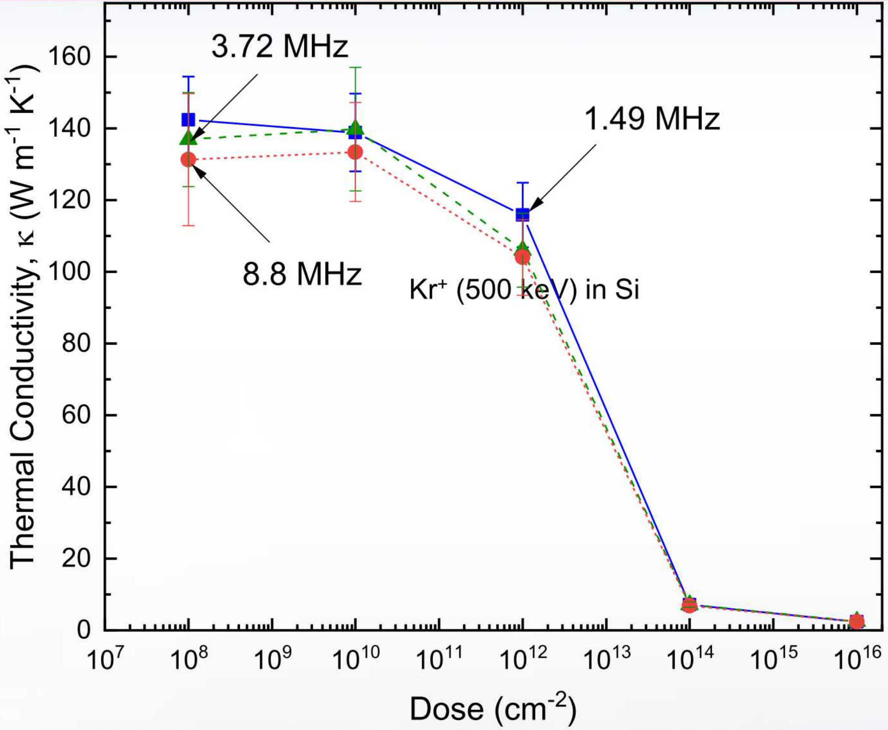
  I. Beyond strain, and implanted point defects can also produce unique defect structures

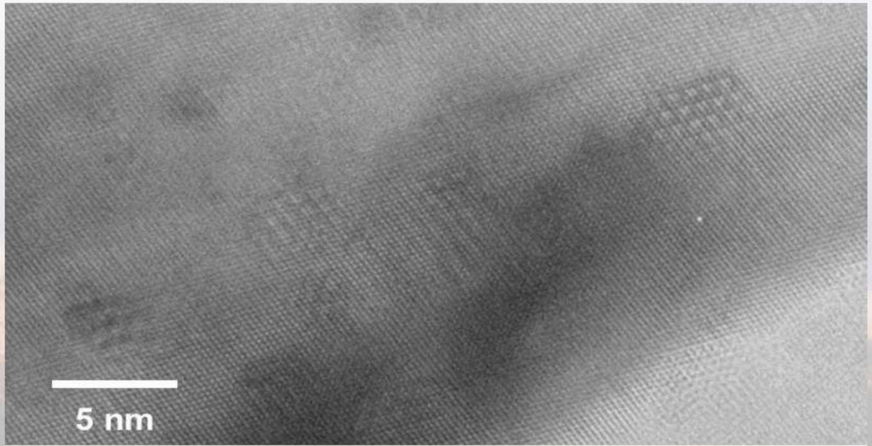
  II. At high enough doses, implanted noble gas ions will cluster into crystalline regions

1) Crystalline gas regions can be created through implantation of noble gases

2) Nearly perfect Lenard-Jones example

3) Greatly reduce thermal conductivity



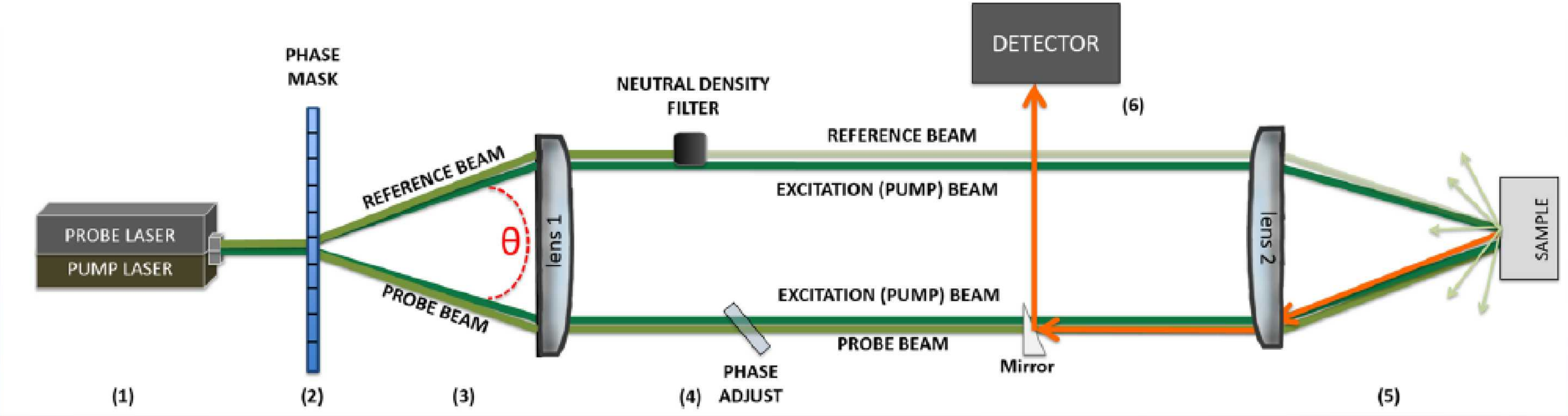


Movement Toward In-situ Measurements

Collaborator: Cody A. Dennett and Mike P. Short

Transcript Grating Spectroscopy (TGS)

I. **Properties**
   I. Fewer required optics
   II. Potential for in-situ measurements
   III. Faster results



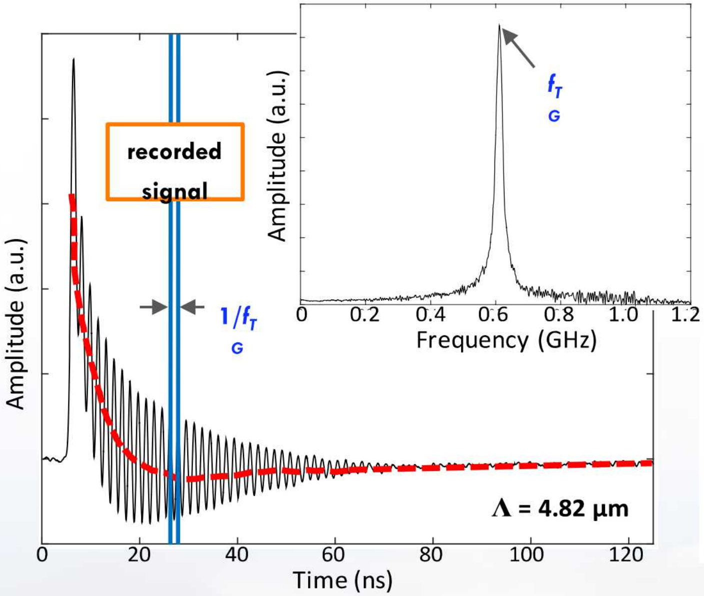
Short et al, *JOM,* **67**, 1840 (2015)

TGS is a faster technique than TDTR, permitting for the possibility of in-situ measurement

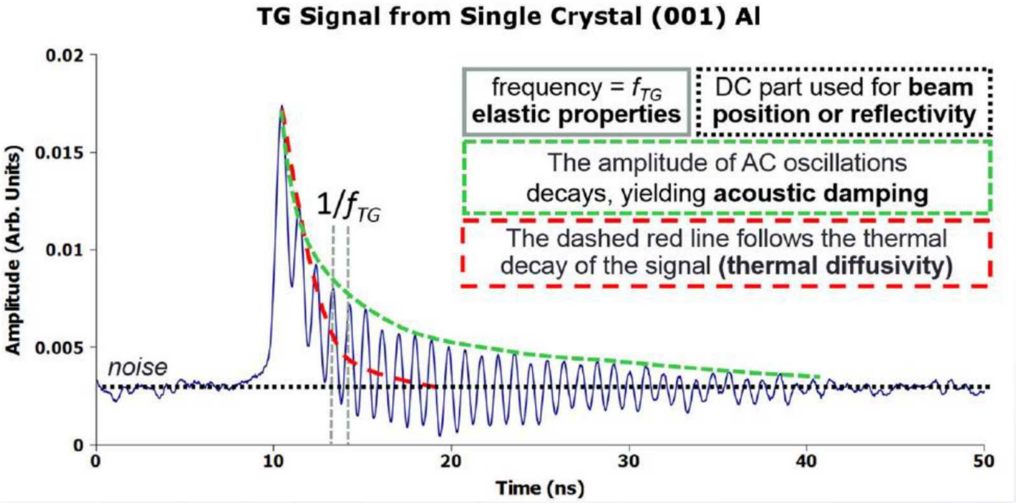
What does TGS Measure?

Collaborator: Cody A. Dennett and Mike P. Short

TGS Signal Analysis



Dennett et. al., *PRB*, **94** (2016)



Short et. al., *JOM*, **67**, 1840 (2015)

Acoustic oscillations return elastic mechanical properties, the grating decay returns thermal properties

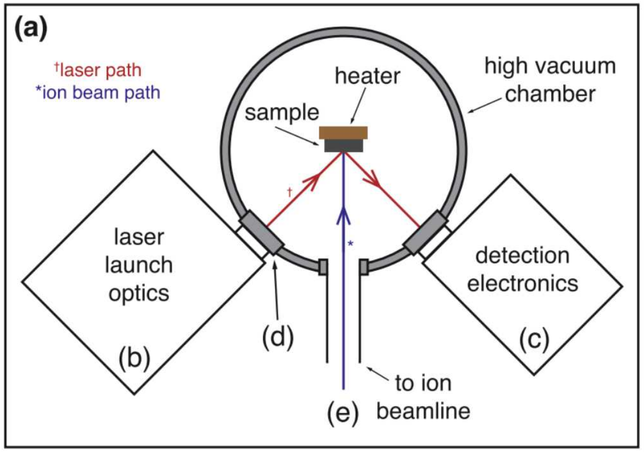
Image or associated Citation

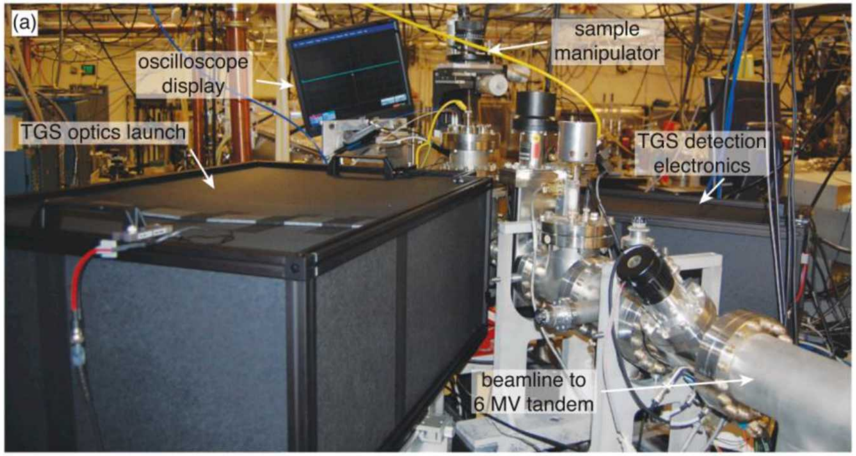
In-situ Experiments with TGS

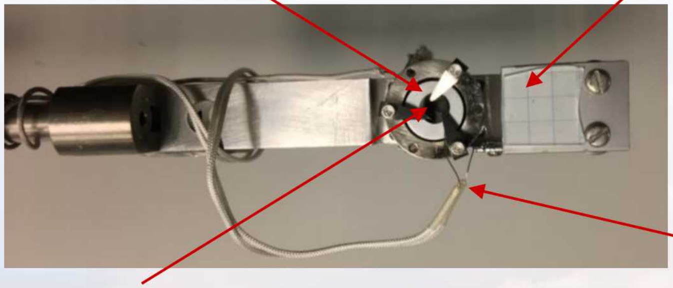
Collaborator: Cody A. Dennett and Mike P. Short

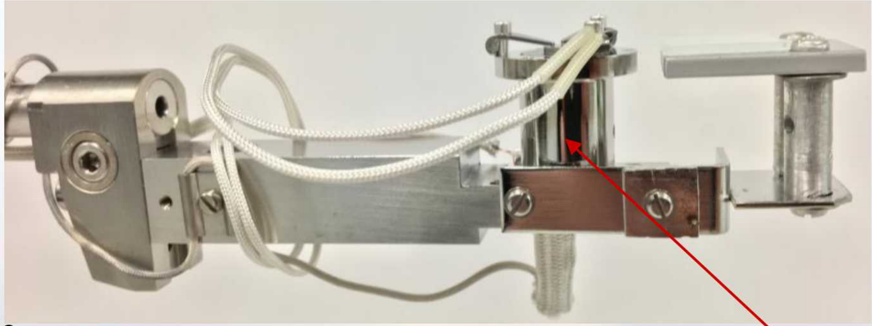
I^{3}TGS at Sandia National Laboratories

Dennett et. al., *Nucl. Instrum. Meth. Phys. Res. B* 440 (2019)









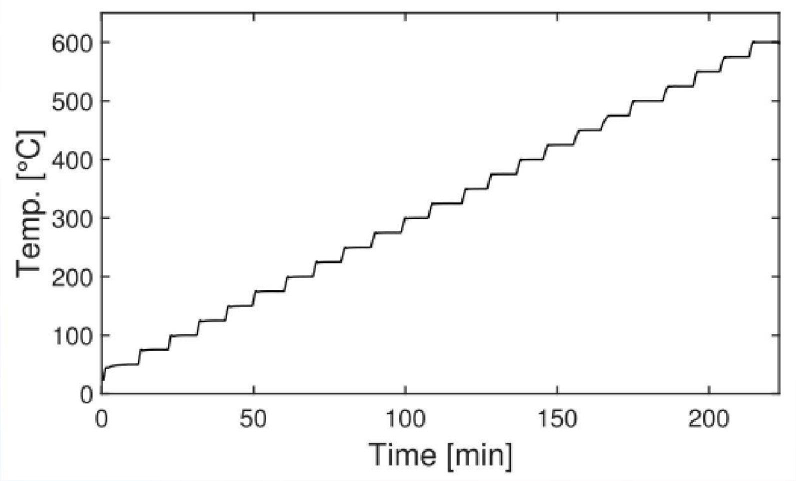
Due to the compact nature of the TGS optical design, an in-situ TGS system has been integrated into Sandia's Ion Beam Laboratory

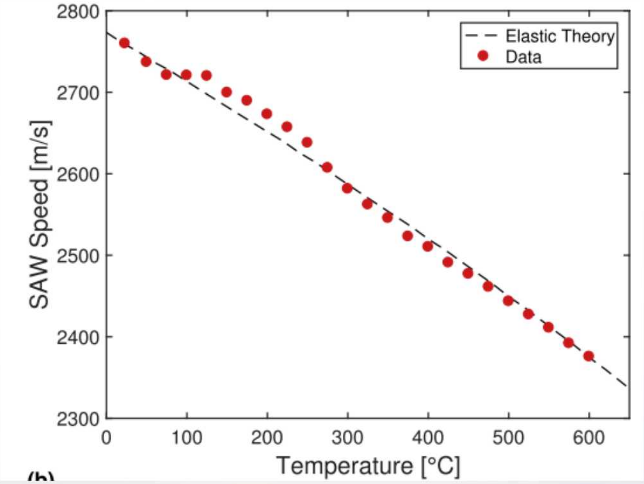
Proof of Concept: Static High T Tests

Collaborator: Cody A. Dennett and Mike P. Short

Static temperature test

I. **{001} Single crystal Nickel**
  I. Chamber pumped to 4.4E-6 Torr
  II. Sample heated from 23 to 600 C (25 C increments)
  III. At each temp., SAW speed measured with TGS
  IV. Data compared to literature calculations





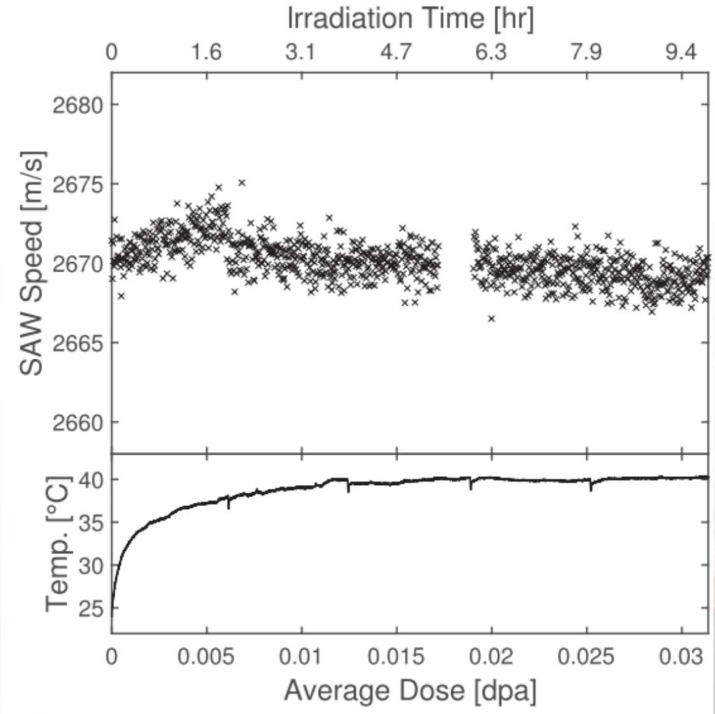
Dennett et. al., *Nucl. Instrum. Meth. Phys. Res. B,* **440**, 126 (2019)

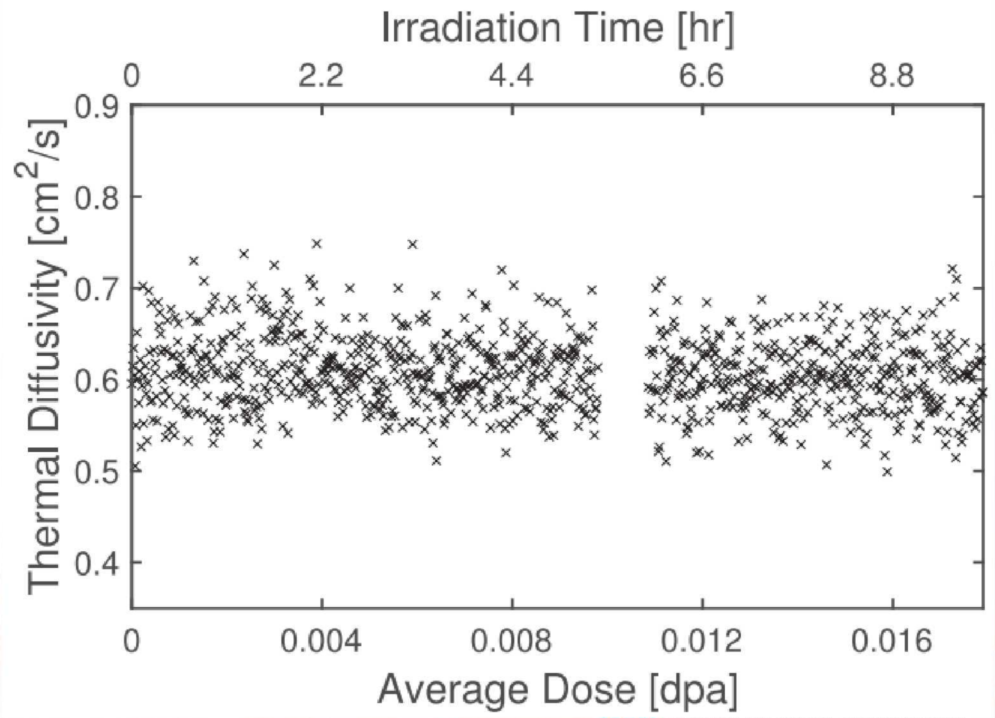
Agreement of the SAW data with literature-based calculations demonstrates temperature stability of the beamline end station

In-situ TGS for Light Ion Exposure

Light Ion Implant

I. **3.7 MeV He⁺ into {001} W crystal**
    I. High interest in W in radiation community due to its potential for fusion applications
    II. Total fluence: 6.7E16 cm^{-2} over 10 hrs
    III. Slow temperature rise due to ion beam heating
    IV. No discernable trends in data (SAW speed, thermal diffusivity)
        I. He bubble formation does not occur until 300 – 3000 appm





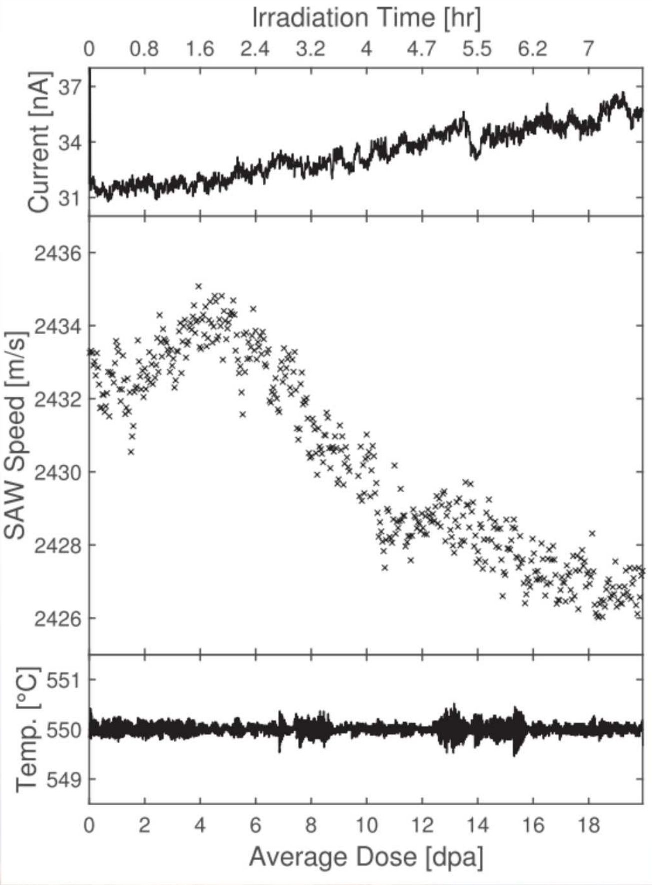
Dennett et. al., *Nucl. Instrum. Meth. Phys. Res. B,* **440**, 126 (2019)

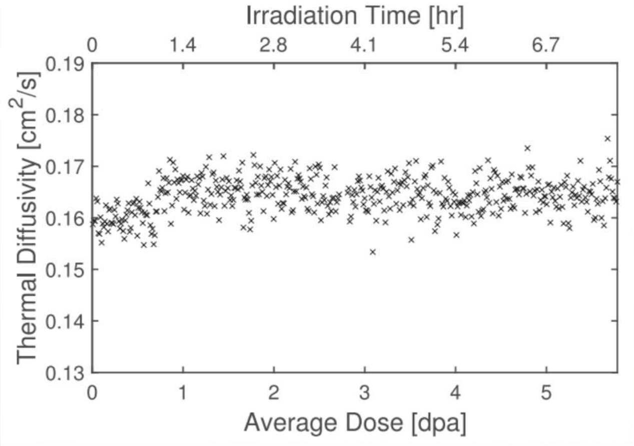
In-situ TGS for Heavy Ion Exposure

Collaborator: Cody A. Dennett and Mike P. Short

Heavy Ion Implant

I. **31 MeV Ni^{5+} into {001} Ni crystal**
  I. Important to many engineering alloys
  II. Has displayed void swelling under self-ion irradiation
    I. Sample heated to 550 C to promote void swelling
  III. SAW speed decreases with dose
    I. Consistent with ex-situ Cu results; material is softened
  IV. Thermal diffusivity remains constant
    I. Defect scattering offset by T dependence of e-e, ep scattering





Dennett et. al., *Nucl. Instrum. Meth. Phys. Res. B,* **440**, 126 (2019)

In-situ TGS can be used to detect the onset of void swelling in irradiated metals

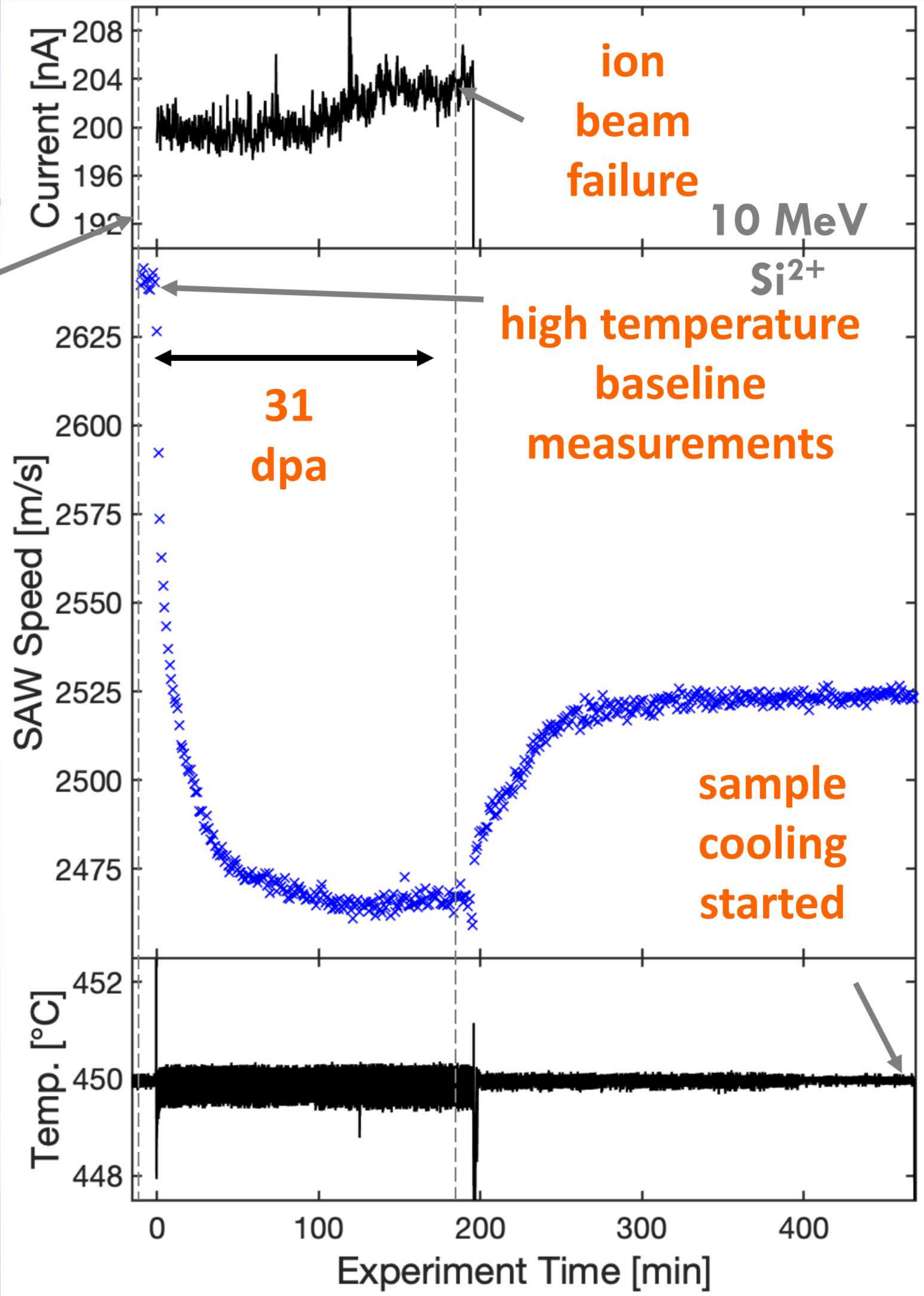
Unexpected *In situ* Results

Collaborator: Cody A. Dennett, Jeff Aguiar, and Mike P. Short

refractory multiple principle element alloy:

13Cr-7Al-23Zr-30Mo-24Nb-4Ta

***In situ* means interesting failures that raise more questions on the materials response to radiation!**



ion beam on sample

ion beam failure

high temperature baseline measurements

31 dpa

sample cooling started

Next Steps…

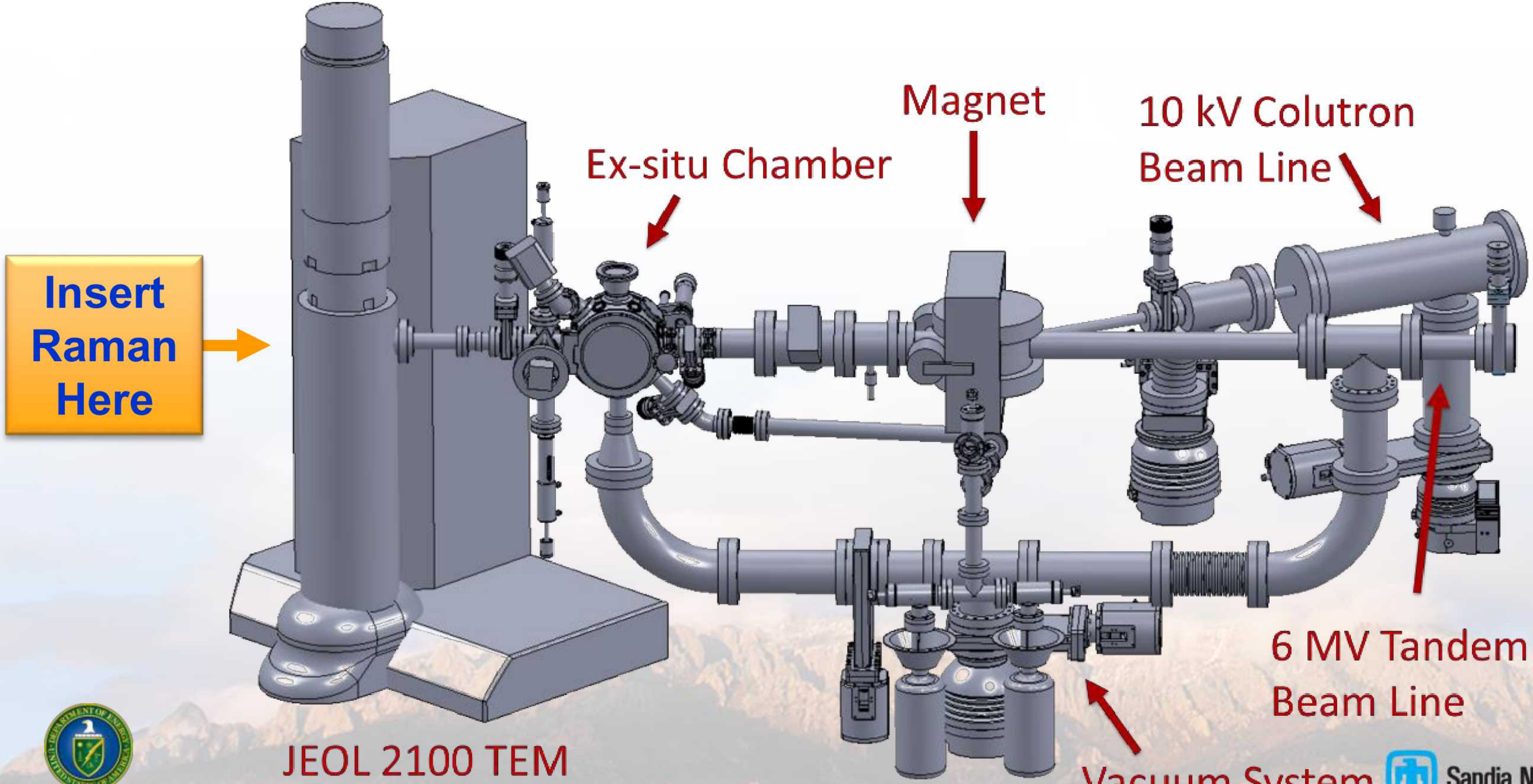
1) Investigate Larger Sample Set

I. Both through passive acoustic listening and laser based pump-probe techniques

2) Add Raman to I^{3}TEM

I. Following on approach from R. Sharma at NIST (TEM) and L. Beck at CEA Saclay (SEM)

II. Add a multi-laser Raman system to the In-situ Ion Irradiation TEM



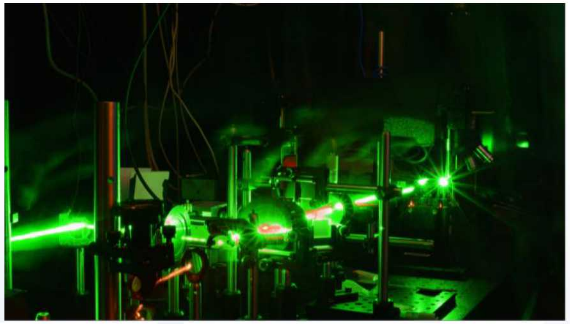
Summary

I. **Intersection of particle accelerators and laser metrologies is providing new avenues of study**
    I. Use listening tools o investigate solid-radiation interactions

II. **AE is a quick and robust technique to identify catastrophic failure.**

III. **TDTR provides an excellent technique to probe thermal conductivity**

IV. **I^{3}TGS offers a means of in-situ measurement of structural properties**



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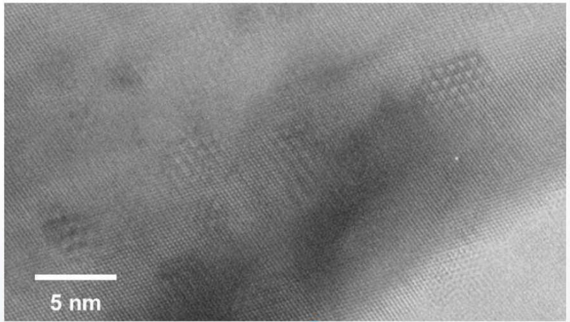

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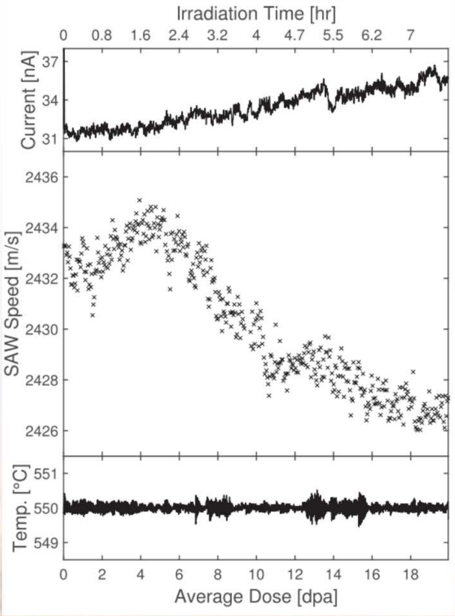


Collaborators:

- Sandia: Daniel Buller, Thomas Beechem, Chris Saltonstall, Elbara Ziade, R.C. Choens, C.A. Taylor, N.M. Heckman, M.D. Ingraham, D. Robinson, B.L. Boyce,
- External: Ethan Scott, Cody Dennett, Michael Short, Patrick Hopkins, Mark Goorsky, Keivan Esfarjani, Jeffrey Braun, Christina Rost, John Gaskins, Mehrdad Fazli, Claire Ganski, Chao Li, Tingyu Bai, Yekan Wang

Sandia’s User and Position Oppurtunities

D. Hanson, W. Martin, M. Wasiolek

www.cint.lanl.gov

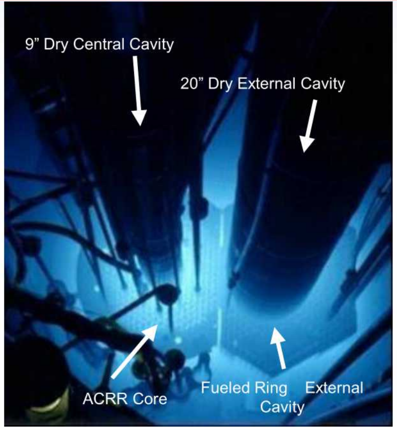
- Spring and Fall proposals for 18 months
- Rapid Access proposal anytime for 3 months

Core Facility - SNL

Gateway Facility - LANL

www.nsuf.inl.gov

- Three proposal a year for 9 months



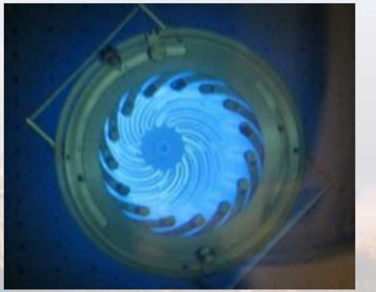
Position Oppurtunities at:

https://www.sandia.gov/careers/

Post-doc = 671121

Grad Student = 670865

Undergrad Student = 670864



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