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Phonon scattering effects in the thermal conductivity reduction of ion irradiated diamond

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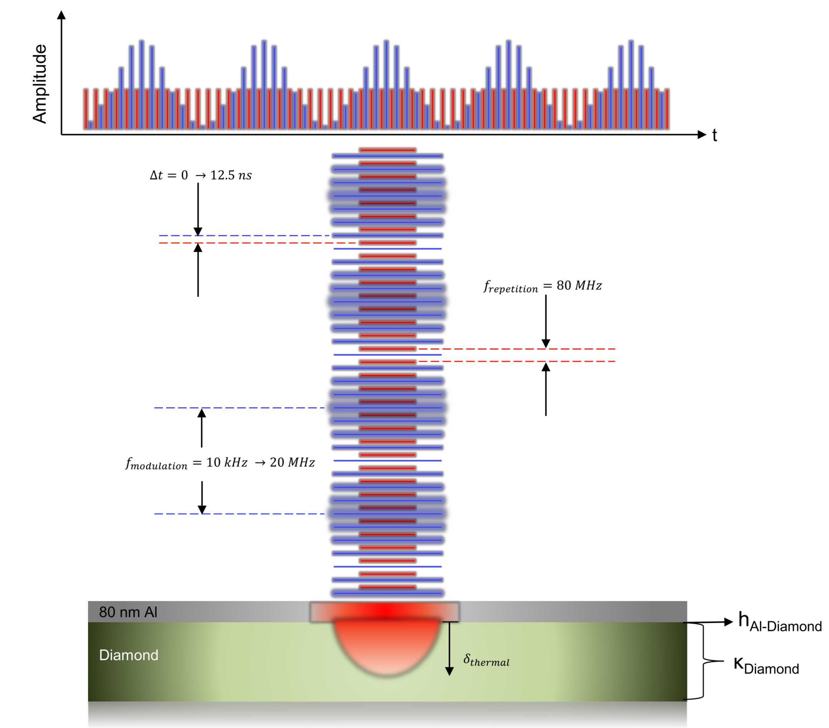
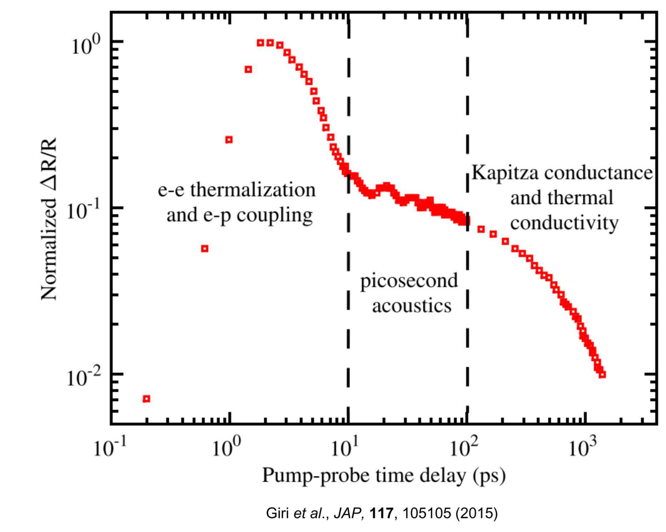
Synopsis

- We implant polycrystalline diamond substrates with ions of C^+, N^+, and O^+. Samples were implanted with a beam energy of 16.5 MeV using a 6 MV Tandem Van de Graaff Pelletron Accelerator with fluences ranging from $4(10^{14})$ to $4(10^{16})$ cm^{-2}.
- The change in mass amongst implant ions offers an avenue to exploit the effect of perturbing mass differences in the defect-scattering term of popular phonon scattering models.
- Time-domain thermoreflectance is used to measure an increase in thermal boundary conductance as well as orders of magnitude reduction in thermal conductivity
- The damage in the samples is characterized through x-ray diffraction as well as scanning transmission electron spectroscopy, which is contrasted against damage predicted with TRIM simulations
- A Klemens model for phonon defect scattering is applied to the thermal conductivity, providing insight into the magnitude of the scattering coefficient for each ion

Time-Domain Thermoreflectance (TDTR)

Non-contact, optical pump-probe technique used to measure material thermal properties



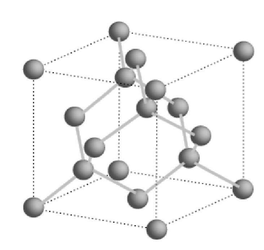


Pump and probe pulses arrive at different times:
- Pump pulse heats the sample
- Probe pulse measures the change in temp at different times after the heating event.

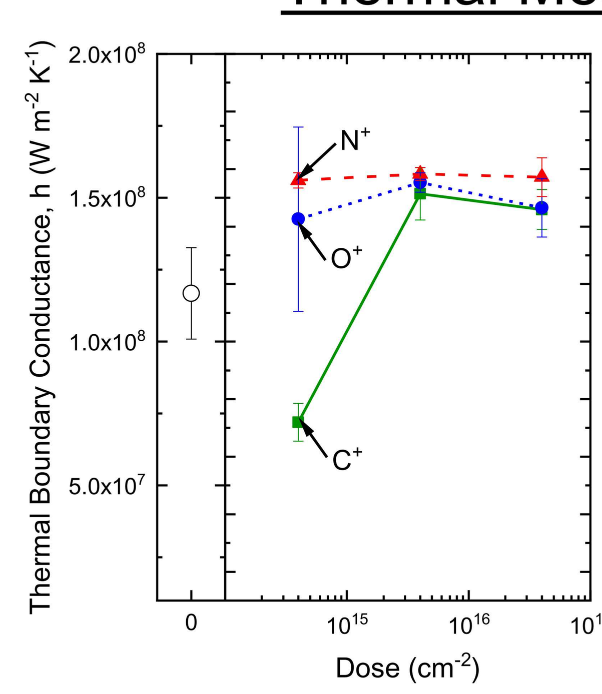
Application of Diamond

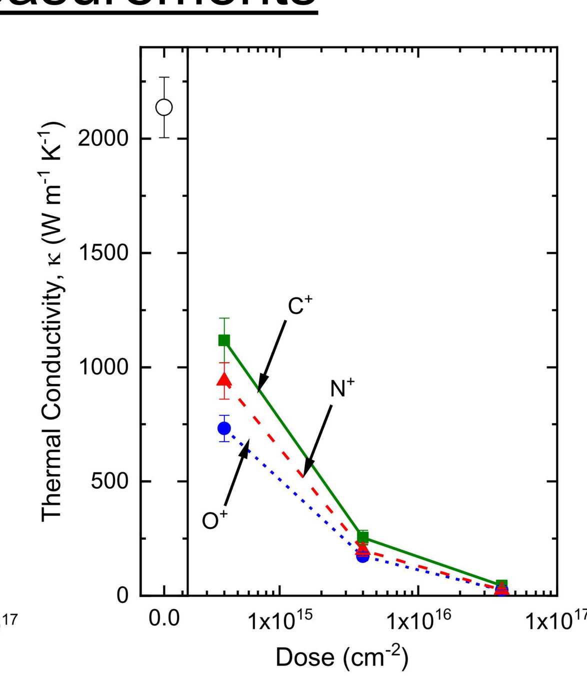
Material	Thermal Conductivity κ (W m-1 K-1)	Electrical Resistivity ρ (Ω cm)	Elastic Modulus E (GPa)
Diamond	2000	10^{13} - 10^{18}	1050 -1210
Silicon	140	0.1-60*	140- 180
Al_2O_3	35	$>10^{18}$	330 - 400
SiC	330 - 400	10^2 – 10^6*	400

- Precision machining
  - Metal processing
- Optics
  - Superlative transmission UV-RF
  - Elimination of thermal lensing
- Sensors
  - Chemically inert
  - Low leakage current
  - Minimal temperature dependence
- Electronics*
  - Thermal management
    - AlGaN/GaN-on-diamond
  - Radiation detection



Thermal Measurements





- Overall increase in thermal boundary conductance observed as a result of irradiation. Similar results have been demonstrated in single crystal silicon
- Decreases in thermal conductivity span two orders of magnitude >2,000 → ~20 W m^{-1} K^{-1}

Thermal Conductivity

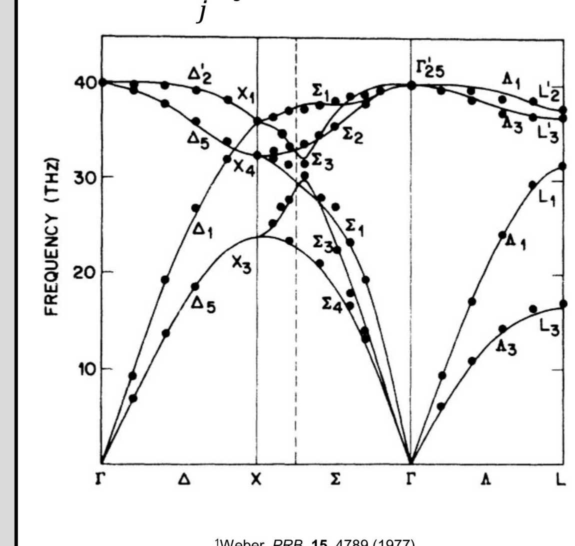
- To gain insight into phonon scattering, Thermal conductivity modeled from a semiclassical kinetic theory approach
- Phonon dispersion taken from literature[1]

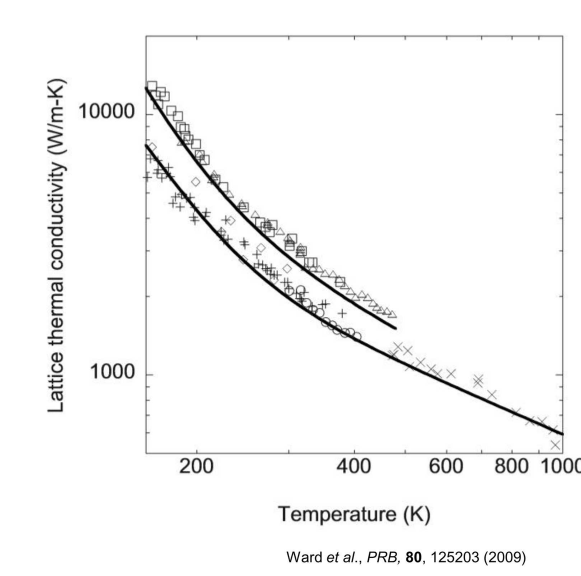
$$\kappa = \frac{1}{3}C_v v l = \frac{1}{3}C_v v^2 \tau = \frac{1}{6\pi^2}\sum_j \int \hbar\omega_j(k)k^2 \frac{\partial f}{\partial T} v_j^2(k)\tau_j(k)dk$$

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

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

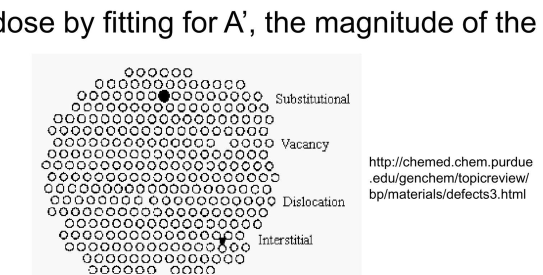
$$\tau_{Imp}^{-1} = A\omega(k)^4$$





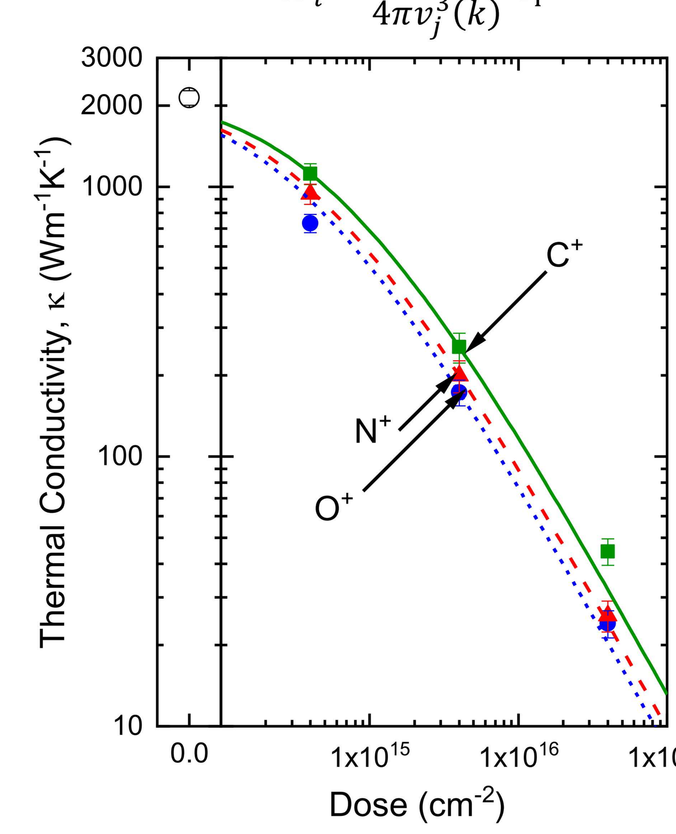
- An additional impurity scattering term is used to model the reduction in thermal conductivity as a function of irradiation dose by fitting for A', the magnitude of the defect scattering term

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



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

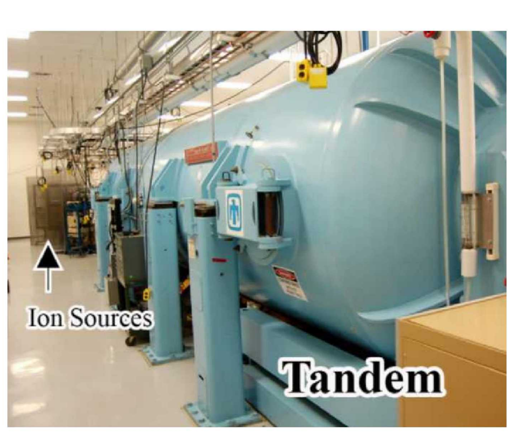
$$A'_i = \frac{\delta^3}{4\pi v_j^3(k)}\Gamma_i$$

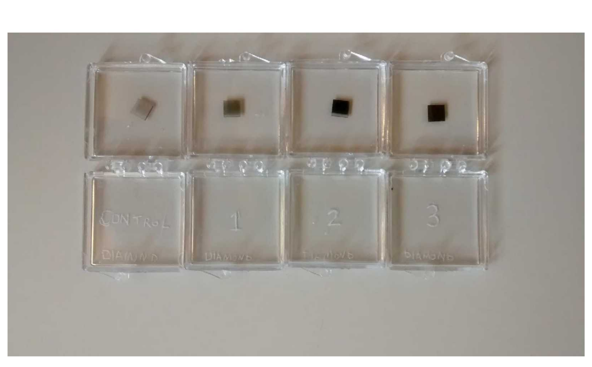


- Good agreement is found between the Klemens model for phonon defect scattering and experimental data
- The magnitude of the scattering coefficients are compared for all ions, and are found to trend with increasing implant mass

i	Percent difference (A'_i,A'_C)	Percent difference (M_i,M_C)
N^+	6.00	15.34
O^+	8.47	28.48

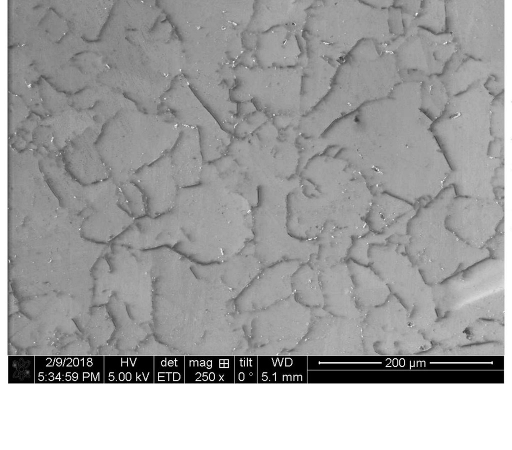
Sample Preparation and Characterization

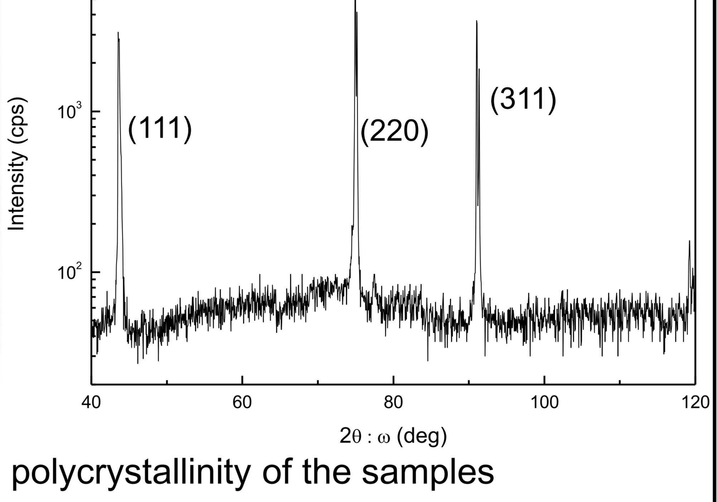




Irradiation Details	
Ion	C+, N+, O+
Energy (eV)	$16.5(10^6)$
Dose (cm^{-2})	$4(10^{14})$, $4(10^{15})$, $4(10^{16})$
Accelerator	6MV Tandem Van de Graaff-Pelletron
Purpose	mass impurity scattering

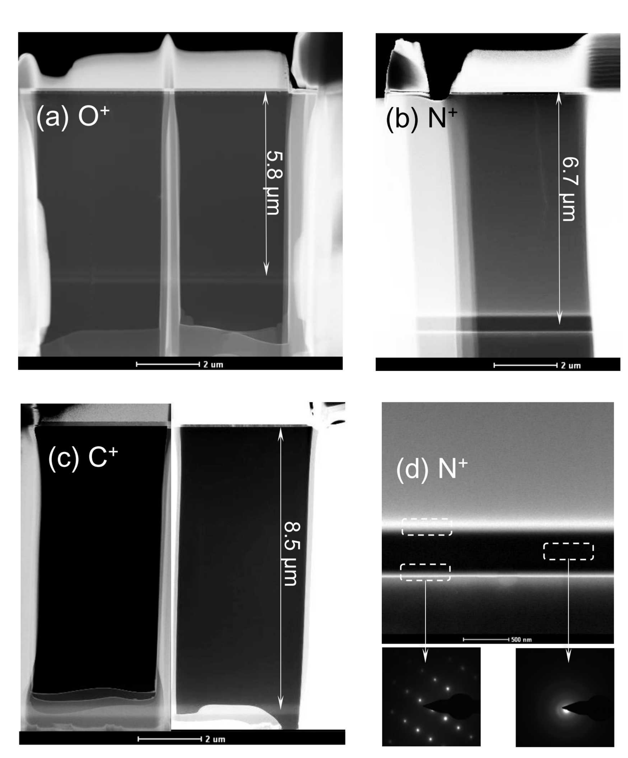
TDTR Preparation	
Sonication	isopropanol, acetone, methanol
O_2 plasma (min)	30
Al deposition	80 nm (e-beam evaporator)

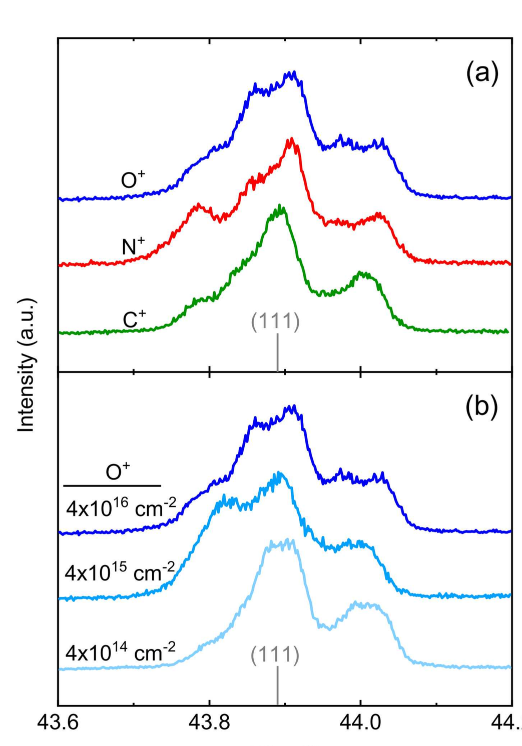


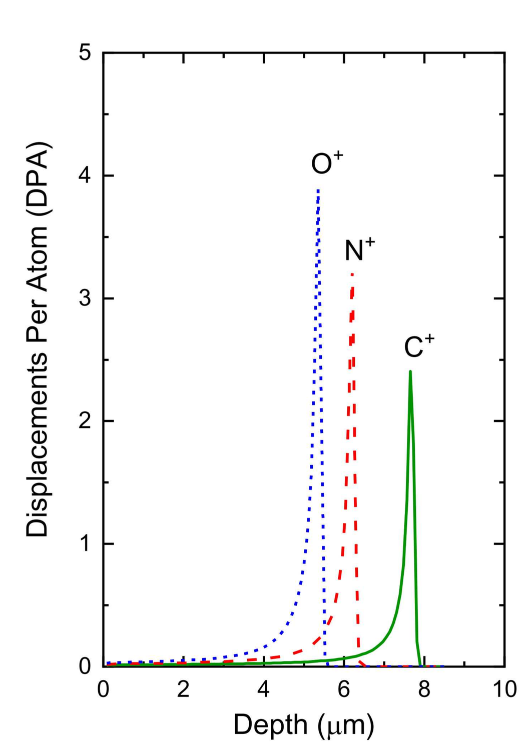


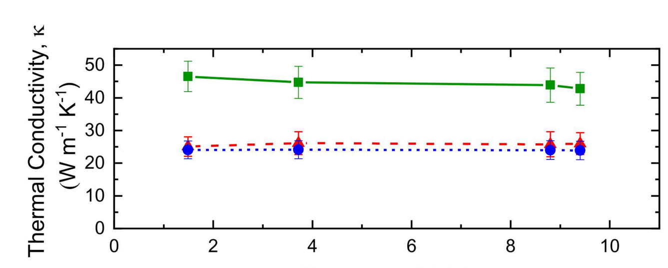
- STEM and XRD confirm polycrystallinity of the samples
- High defect concentration produces a change in color, yielding varying shades of green

Damage Characterization









Ion	δ_{TRIM} (μm)	δ_{TEM} (μm)
O^+	5.355	5.8
N^+	6.205	6.7
C^+	7.65	8.5

- TRIM software used to predict damage profiles for each ion implanted at an energy of 16.5 MeV
- STEM employed to measure the actual projected range at the highest dose
- Lack of diffraction with SAD confirms amorphicity in the regions of highest damage; diamonds maintain polycrystalline structure outside of these bands
- Broadening of the (111) peak in 2θ:ω indicates variation in interplanar spacing, suggestive of strain

Acknowledgements

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