In situ observation of short- and long-timescale material property evolution under extreme conditions

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Collaborators
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Khalid Hattar (SNL)
Michael P. Short (MIT)
material performance is challenged under extreme environments

- temperature
- corrosives
- radiation
- pressure
- fatigue
material performance is challenged under extreme environments

- temperature
- corrosives
- radiation
- pressure
- fatigue
micro-scale

Pure Cu
35 MeV Cu$^{6+}$
400°C
micro-scale

5 µm

Pure Cu
35 MeV Cu$^{6+}$
400°C

macro-scale

in situ characterization

material structure

material properties
in situ characterization

material structure

material properties
This arrangement permits validating electrical components. Allowable by the physical connection and final ion beam characterization.

To address these concerns, an ion beam profile monitor is used to characterize both ion beams at the micro-Raman setup. The micro-Raman setup can be easily switched from one system to another. This facility aims at performing point acquisition but also two-dimensional (2D) maps and depth scans, before and after ion irradiation. The triple-beam chamber allows single, dual or triple-beam irradiation.

The micro-Raman setup is connected to the triple-beam chamber. A 100-mW, frequency-doubled Nd: YAG (532 nm) laser, with four power filters, is used as the excitation source for Raman spectroscopy. A 57-cm extension, an objective with 1000 and 2400 lines/mm and edge filters, allows 1 cm × 1 cm coverage of the sample. A scanner unit able to spread the beam homogeneously over an area of 1 cm × 1 cm × 0.5 mm is equipped with a raster scanner, allowing 50, 100, 200, and 500 times magnification (1800 and 2400 lines/mm) and edge filters, allowing 1 cm × 1 cm × 0.5 mm to obtain 16 power levels (from 0.00005 to 100%).

The Raman system connected to the triple-beam chamber with an incidence angle of 15°, is equipped with a raster scanner, allowing 1 cm × 1 cm × 0.5 mm × 50, 100, 200, and 500 times magnification (1800 and 2400 lines/mm) and edge filters, allowing 1 cm × 1 cm × 0.5 mm to obtain 16 power levels (from 0.00005 to 100%).


S. Miro et al., *J. Raman Spectrosc.* **47** (2016)
in situ characterization

material structure

material properties
Transient Grating Spectroscopy (TGS)
Pump Pulse

Transient grating spectroscopy is a technique which measures thermal-mechanical properties by inducing and monitoring surface acoustic waves.
Figure 3-4: Sample results from transient grating excitation with $\lambda = 4.82 \, \mu m$ along the $(100)$ direction of {001} oriented single crystal aluminum. The SAW response is superimposed over the decaying thermal grating as a function of time. The inset shows the filtered power spectrum of the response with a clear peak at the SAW frequency. Figure sourced from [35].

3.2 Amplitude and phase grating thermal transport

To use the TGS technique to monitor the thermal transport properties of bulk materials under investigation, we must first construct an appropriate model for both the surface temperature profile and the surface displacement profile as a function of time. Consider a two-dimensional semi-infinite half-space defining a surface (at $z = 0$) on which a periodic transient grating will be projected.

We define the positive $\hat{z}$ direction into surface. The surface temperature profile resulting from a $-\delta$-function grating excitation can be found by solving the thermal diffusion equation:

$$
c \nu \frac{\partial T}{\partial t} = k_x \frac{\partial^2 T}{\partial x^2} + k_z \frac{\partial^2 T}{\partial z^2} + Q(x, z, t),
$$

where $c$ is density, $\nu$ is the heat capacity, $T$ is the temperature, $k_x$ and $k_z$ are the thermal conductivity components parallel and perpendicular to the surface, respectively, and $Q(x, z, t)$ is the absorbed heat density [24, 22]. This equation can be solved for an infinite periodic surface excitation of the form:

$$
Q(x, t) = Q_0 \cos(qx) \delta(t),
$$

where $Q_0$ is a laser intensity, $q$ is the grating wave vector as defined in Eq. (3.1), and $\delta(t)$ is the Dirac delta function, by using a Fourier transform technique for both spatial and temporal components of the response [22]. Appropriate boundary conditions for the problem are given by:

$$
\frac{\partial T}{\partial z}(z = 0) = 0 \quad T(z = 1) = T_{\text{bulk}},
$$

(3.10)
Figure 3-4: Sample results from transient grating excitation with $\lambda = 4.82 \mu m$ along the (100) direction of {001} oriented single crystal aluminum. The SAW response is superimposed over the decaying thermal grating as a function of time. The inset shows the filtered power spectrum of the response with a clear peak at the SAW frequency. Figure sourced from [35].

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$$\rho c v \frac{\partial T}{\partial t} = k_x \frac{\partial^2 T}{\partial x^2} + k_z \frac{\partial^2 T}{\partial z^2} + Q(x, z, t),$$  

(3.8)

where $\rho$ is density, $c_v$ is the heat capacity, $T$ is the temperature, $k_x$ and $k_z$ are the thermal conductivity components parallel and perpendicular to the surface, respectively, and $Q(x, z, t)$ is the absorbed heat density [24, 22]. This equation can be solved for an infinite periodic surface excitation of the form

$$Q(x, t) = Q_0 \cos(qx) \delta(t),$$  

(3.9)

where $Q_0$ is a laser intensity, $q$ is the grating wave vector as defined in Eq. (3.1), and $\delta(t)$ is the Dirac delta function, by using a Fourier transform technique for both spatial and temporal components of the response [22]. Appropriate boundary conditions for the problem are given by

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(3.10)

$\Lambda = 4.82 \mu m$
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$$c_v \frac{\partial T}{\partial t} = k_x \frac{\partial^2 T}{\partial x^2} + k_z \frac{\partial^2 T}{\partial z^2} + Q(x, z, t),$$

where $c_v$ is density, $T$ is the temperature, $k_x$ and $k_z$ are the thermal conductivity components parallel and perpendicular to the surface, respectively, and $Q(x, z, t)$ is the absorbed heat density [24, 22].

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$$\frac{\partial T}{\partial z}(z = 0) = 0 \quad T(z = 1) = T_{\text{bulk}}.$$
<table>
<thead>
<tr>
<th>material</th>
<th>single crystal {111} Cu</th>
</tr>
</thead>
<tbody>
<tr>
<td>ion species</td>
<td>Cu(^{6+})</td>
</tr>
<tr>
<td>ion energy</td>
<td>35 MeV</td>
</tr>
<tr>
<td>temperature</td>
<td>400°C</td>
</tr>
<tr>
<td>spot size</td>
<td>0.19 cm(^2)</td>
</tr>
<tr>
<td>beam current</td>
<td>100-250 nA</td>
</tr>
<tr>
<td>target doses</td>
<td>0, 5, 10, 30, 50, 100 dpa</td>
</tr>
</tbody>
</table>

SRIM Dose Profile

35 MeV Cu\(^{6+}\) in Cu at 400°C

dislocation-small defect cluster interactions†

volumetric swelling -4.5%


HAADF STEM* swelling confirmation

Dose (dpa) vs. Depth (μm)

5 dpa  10 dpa  30 dpa  50 dpa  90 dpa

*high angle annular dark field scanning transmission electron microscopy

in situ ion irradiation TGS ($^{13}$TGS) beamline at the Sandia Ion Beam Lab
Dennett et al., *NIMB* 440 (2019) 126-138

$I^3$TGS target chamber
long- and short-timescale defect effects in Ni-based solid solution alloys

- high strength
- thermal stability
- wear resistance
- corrosion resistance
- irradiation tolerance

bright-field TEM of alloys irradiated with 1.5 MeV Ni⁺ to 60 dpa peak at 500°C
<table>
<thead>
<tr>
<th>equiatomic composition</th>
<th>single/polycrystal</th>
<th>source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni</td>
<td>SC {001}</td>
<td>commercial</td>
</tr>
<tr>
<td>NiFe</td>
<td>SC {001}</td>
<td>ORNL</td>
</tr>
<tr>
<td>NiCoCr</td>
<td>SC {001}</td>
<td>ORNL</td>
</tr>
<tr>
<td>NiFeCoCr</td>
<td>SC {001}</td>
<td>ORNL</td>
</tr>
<tr>
<td>NiFeCoCrMn</td>
<td>polycrystal</td>
<td>ORNL</td>
</tr>
</tbody>
</table>

*in situ* long-timescale irradiation test matrix
### in situ long-timescale irradiation test matrix

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<thead>
<tr>
<th>equiatomic composition</th>
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<td>ORNL</td>
</tr>
</tbody>
</table>

*pre-irradiation TGS characterization used to identify acoustic polarization for in situ testing*
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>surface angle</td>
<td>(&lt;100&gt;{001&gt;)</td>
</tr>
<tr>
<td>ion species</td>
<td>Ni(^{5+})</td>
</tr>
<tr>
<td>ion energy</td>
<td>31 MeV</td>
</tr>
<tr>
<td>temperature</td>
<td>550°C</td>
</tr>
<tr>
<td>TGS wavelength</td>
<td>4.55 (\mu) m</td>
</tr>
<tr>
<td>peak dose</td>
<td>60 dpa</td>
</tr>
<tr>
<td>peak dose rate</td>
<td>1.6-1.8 (\times 10^{-3}) dpa/s</td>
</tr>
<tr>
<td>measurement time</td>
<td>35 sec</td>
</tr>
<tr>
<td>measurement interval</td>
<td>60 sec</td>
</tr>
<tr>
<td>exposure time per sample</td>
<td>9.5-10.5 hours</td>
</tr>
</tbody>
</table>

void swelling at high temperatures and high doses

31 MeV Ni\(^{5+}\) in Ni at 550°C
Pure Ni
NiFe

- [Graph 1: Thermal Diffusivity vs. Peak Dose]
- [Graph 2: Irradiation Time vs. Current, SAW Speed, Temp.]
NiCoCr

- Graph showing thermal diffusivity vs. peak dose.
- Graph showing irradiation time vs. current and SAW speed.
- Graph showing temperature vs. peak dose.
NiFeCoCr
NiFeCoCrMn
ion beam direction

Ni
NiFe
NiCoCr
NiFeCoCr
NiFeCoCrMn

500 nm

dose map (dpa)

0 1 2 3 4 5 6 µm

dose

0 5 10 15 20 25 30 35 40 45 50 55 60
at temperature, ion beam off

at temperature, ion beam on

`instantaneous` defect generation affects mechanical properties
<table>
<thead>
<tr>
<th>surface angle</th>
<th>&lt;100&gt;{001}</th>
</tr>
</thead>
<tbody>
<tr>
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<td>ion energy</td>
<td>31 MeV</td>
</tr>
<tr>
<td>temperature</td>
<td>500°C</td>
</tr>
<tr>
<td>TGS wavelength</td>
<td>4.55 µm</td>
</tr>
<tr>
<td>dose rates</td>
<td>[1, 2, 5, 10, 20] x 10^{-4} dpa/s</td>
</tr>
<tr>
<td>measurement interval</td>
<td>~30 sec</td>
</tr>
<tr>
<td>annealing time between impulses</td>
<td>~30 min</td>
</tr>
</tbody>
</table>

instantaneous defect effects at varying dose rates

![Graph showing SAW Displacement and Ion Damage](image-url)

31 MeV Ni$^{5+}$ in Ni at 550°C

**Magnitude [a.u.]**

**Depth [µm]**
example current and temperature records for NiCoCr

beam current lowered in steps between impulse experiments

higher beam heating and larger temperature fluctuations during high flux experiments
Pure Ni

transient defect populations stiffen the elastic modulus, inversely correlated with dose rate

background defect accumulation continually evolving and not fully annealing
NiFe

transient defect populations soften/reduce the elastic modulus, magnitude directly correlated with dose rate

blue → dose rate high to low
red → dose rate low to high
NiCoCr

transient accumulation time longer than for other alloys

elastic constant reduction greatest observed of any alloy chemistry studied
NiFeCoCr

Initial lattice stiffening is irreversible, softening observed at identical dose rates following saturation.
short-timescale defect accumulation observations and question

stiffening vs. softening in different alloys
defect type variation?

limitations of subsequent irradiation impulses
underlying background evolution?
higher temperatures needed for annealing?
in situ means interesting failure

13Cr-7Al-23Zr-30Mo-24Nb-4Ta
refractory multiple principle element alloy
• *in situ* ion irradiation TGS beamline available for use at Sandia National Labs

• thermoelastic material properties explored directly *in situ* under extreme conditions at both short and long timescales
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